THE KELLOGG COAL GASIFICATION PROCESS

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I.   INTRODUCTION

The Kellogg Molten Salt Coal Gasification Process represents a unique approach to the problem of coal gasification. The molten sodium carbonate system used for heat supply and catalysis in the Kellogg Coal Gasification Process provides the following advantages. The strong catalytic action for the steam-carbon reaction and coal combustion reaction makes it possible to carry the overall gasification to essentially complete conversion of coal leaving very little fuel to be rejected with the ash. Heat can be supplied by air or oxygen combustion of coal or char from the molten salt, and, in the case of oxygen, it is possible to keep the combustion products separated from the synthesis gas. This has the advantage of keeping sulfur oxides out of the flue gas stream and, in addition, it eliminates carbon oxide dilution of the synthesis gas stream.

Examination of the basic process chemistry and the effect of pressure on the process economics has shown that the Kellogg process is improved by operation at 1200 psia rather than at 400 psia as in the original concept. Operation at high pressure and the separation of the gasification zone from combustion zone makes it possible to take full advantage of the catalytic effect of the molten salt on the direct production of methane from synthesis gas in the gasification step. This is important because it will minimize the heat supply requirement thereby yielding improved economics.

In this process, steam and fine coal are injected continuously into a molten salt bath where they react to form synthesis gas according to Reaction (1):

$$C + H_2O + heat \rightarrow H_2 + CO \quad\quad\quad (1)$$
$$\text{(coal) (steam)} \quad\quad \text{(synthesis gas).}$$
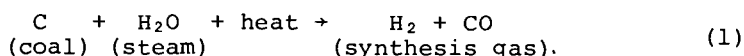
The necessary heat of reaction is supplied by circulating a heated salt stream. In addition, the molten salt mixture is chosen to catalyze Reaction (1) so that it may be carried out at a relatively low temperature. Savings result from the lower reaction temperature, which minimizes the heat demands of the process. The lower reaction temperature also results in a larger concentration of methane in the synthesis gas, which is a decided advantage in the manufacture of pipeline gas. The result is lower investment and lower operating costs.

Since the coal to be gasified is suspended in a salt melt, physical properties of the coal particles are not as critical as in a fixed-bed, moving bed, or fluidized-bed gasifier, where agglomeration and gas channeling must be avoided. It is expected, therefore, that caking, as well as non-caking, coals can be gasified in a continuous manner. Other fuels which can be gasified include petroleum residual fractions (reduced crude), shale, tar sands and municipal waste (garbage)—a brief review of waste gasification has shown this to be feasible. There is, of course, some economic penalty associated with the higher ash contents of some of these fuels. Moreover, better fuel utilization can be obtained using larger particles—hence less grinding equipment and power consumption—than in a suspension gasifier.

The raw synthesis gas produced by the process can subsequently be converted: to a synthetic pipeline gas by catalytic methanation; to hydrogen by water-gas shift and carbon dioxide removal; or to liquid fuels and chemicals via Fischer-Tropsch synthesis.

The molten salt gasification process development program has proceeded in three general areas which include (1) the basic chemistry of the process (2) the testing of materials of contruction to withstand the corrosive, high-temperature environment and (3) engineering feasibility and economic studies.

The present status of the development is illustrated by the following typical results in these three areas. Review of the development work is not complete since time is not available in this symposium to present all the development work.

II. PROCESS STATUS

A. PROCESS CHEMISTRY

The bench-scale process research work was designed to provide the data required to fix the process operation conditions needed to ensure attractive economics. The bulk of the effort was directed toward determining the effects of the major process variables on the rates of gasification and combustion of a broad range of coal feeds. Other work done included viscosity measurements of various melts, melt expansion as a function of gas flow, measurement of salt carry-over, disposition of sulfur in the melt and the development of a recovery procedure for sodium carbonate while removing ash.

The experimental work on gasification and combustion has been carried out in a reactor consisting of a two-inch I.D. × 26 inch long Inconel pipe enclosed in an electric furnace. Variables which have been tested are coal rank, coal particle size, steam partial pressure, temperature, superficial gas velocity, melt depth and ash and carbon content of the melt.

The activation energies for various ranks of coal have been found to vary from 5300 cal/mole for Renner's Cove lignite to 28,600 for anthracite. The gasification rate was found to increase with the 0.55 power of the steam partial pressure and favorable effects on the gasification rate were also found with increasing gas velocity and ash content. The rate is first order with respect to carbon content of the melt but the gasification rate was found to decrease with increasing melt depth. Further gasification tests are planned in the larger, 1200 psi vessel described in the next section on materials testing.

The combustion rate was similarly found to be first order with carbon content but no effect of air pressure or velocity was found. Finer size of coal particles was found to increase the combustion rate and the addition of 0.5% Na_2S increased the rate by a factor of four. Once again, increasing melt depth was found to reduce the combustion rate. The combustion efficiency to CO_2 has been measured to be greater than 85%. Studies on the disposition of sulfur in the process have shown that the flue gas will contain very little sulfur compounds.

B. MATERIALS TESTING

The molten salt system has presented a severe containment problem. Previous work on corrosion of test specimens has shown that Monofrax A (a fused, cast, high-purity alumina) provides satisfactory corrosion resistance and can serve well as a vessel lining. The corrosion tests have included all the components that will be in the reactor under commercial operating conditions, including coal ash, coal sulfur, the sodium-sulfur compounds in the appropriate state of reduction and oxidation as well as coal particles undergoing gasification. In addition corrosion tests of Monofrax A were conducted with test pieces that were spring loaded to compressive stress levels that would exist in the wall of a full-scale commercial vessel. These tests showed clearly that Monofrax A would resist the attack of the reacting system including design stresses in a completely satisfactory manner.

Vessel designs were carried out in sufficient detail to ensure that dependable vessels could be built using a Monofrax A lining. The mortar in the wall will be frozen salt necessitating proper control of temperature gradients through the wall, and designs incorporating such gradient control were found to be entirely practical. It has become necessary to demonstrate the validity of the conclusions from the corrosion test program and the vessel design work and thus to demonstrate the validity of the design for the containment problem. For demonstration purposes, a corrosion test vessel has been designed.

It is expected that this corrosion test unit will, when commissioned, provide proof of feasibility of the commercial design concept. The test vessel chosen is six inches diameter by six feet long and the heat is supplied by combustion of fuel

to produce a simulated synthesis gas. This permits establishment and demonstration of the temperature gradients essential to the design, and avoids the very high cost of a melt circulation system. The fuel chosen is propane which is burned with air at full process pressure. Steam, hydrogen sulfide and coal ash are added to the system to simulate the "working" salt conditions that would be encountered in actual coal gasification.

III. PROCESS DESCRIPTION

Based on the experimental work carried out on the process, a flow sheet for a plant to produce 262,000,000 standard cubic feet per day of 956 Btu/SCF pipeline gas has been prepared. The gasification section of this plant is shown in Figure 1. This flow sheet design work has had the objective of determining the economic attractiveness of the process; some items (e.g., coal feeding) may be modified on further design work. Ground coal is added to one of the coal lock hoppers and is pressurized with an inert gas or with compressed synthesis gas. Coal is withdrawn continously from the pressurized hopper (while the other hopper is being filled) into to stream of steam at 1500°F and 1250 psia. Steam and coal are injected into the molten salt gasifier where they are heated by intimate contact with the molten salt (sodium carbonate) and react according to Reaction (1) to produce synthesis gas. The synthesis gas leaves the molten salt bath at about 1750°F and 1200 psia, flows through a heat exchanger where the sensible heat of the gas is used to generate steam, is quenched with water and is delivered at 700°F for further processing.

In the molten salt gasifier where Reaction (1) takes place, a circulation of molten salt is used to supply the needed heat. This heat is generated in a separate combustion vessel where some of the coal in the melt is burned with oxygen which heats the melt. Melt circulation is accomplished using the gas lift principle where steam is used to aerate and lift the molten salt from the combustor to the gasifier. Flow from the gasifier back to the combustor is by gravity.

The flue gases which heat the melt by direct contact are generated by combustion of coal with oxygen. Oxygen for this purpose is compressed to 1230 psia and is preheated against the flue gases leaving the combustor. The flue gases flow through the expander which provides the power needed for oxygen compression and then flow to the ash removal section where the CO_2 is used to recover the Na_2CO_3.

It should be noted at this point that the two-vessel, gasifier-combustor system shown in Figure 1 is but one of the possible designs which are being considered. One alternate is a single-vessel system where the oxygen is fed directly to the gasifier. This scheme eliminates the circulating melt but results in reduced methane production in the gasifier and somewhat higher operating costs.

Ash left in the melt by the combustion and gasification of the coal is allowed to build up to a level of 8 weight percent and settled in a quiescent zone to 16 percent ash. A stream of the ash-carbon-sodium carbonate mixture is continuously withdrawn from the gasifier and flows to ash removal where it is processed to separate the ash from the melt. A simplified flow sheet for this ash removal is presented as Figure 2. The melt stream is quenched to 400°F with a solution saturated with sodium bicarbonate at 100°F in the quench tower. Solid melt particles in the resulting slurry are ground to facilitate dissolution of the salt. This stream is then flashed to essentially atmospheric pressure into a holding tank, where sufficient residence time is provided to dissolve the sodium carbonate. The slurry leaving this vessel is filtered to separate the ash and carbon from the solution. This residue is sent to disposal.

The solution leaving the filter flows to a carbonation tower where the sodium carbonate is reacted with carbon dioxide from the gas purification system. The tower operating temperature is about 100°F. At this temperature the sodium bicarbonate concentration exceeds its solubility limit and it is precipitated from solution. The slurry is then filtered, the bicarbonate leaving the filter is calcined to decompose the bicarbonate to carbonate and is returned to the combustor while the solution is recycled to the quench tower.

Raw synthesis gas delivered from the synthesis section is further processed as shown in Figure 3. The gas is shifted to adjust the ratio of hydrogen to carbon monoxide for optimum methanation, and is then purified and finally converted to methane.

IV. ECONOMICS

The equipment needed to carry out the primary process steps just described as well as all auxiliary equipment needed to supply additional services and controls have been designed for a plant to produce 262,000,000 standard cubic feet of 956 Btu/SCF pipeline gas and the plant cost has been estimated.

Estimated capital investment is summarized in Table 1. Offsite Facilities, Section 1100, encompasses all those facilities needed to make the plant completely self-sufficient, such as fire-fighting equipment, buildings to house administrative and supporting personnel, and equipment to supply steam, electric power and cooling water. Total capital investment for the plant is about $149,000,000. This investment is for a "turn key" plant—that is, one which is completely erected and ready for full operation—and includes the contractor's overhead and profit for the complete engineering and construction of the plant.

Estimated gas manufacturing cost and selling price are shown in Table 2. With bituminous coal charged at a nominal $4 per ton and depreciation at 5 percent per year, estimated gas production cost, before any return on investment, is about 52¢/10^6 Btu. Using the AGA-IGT Utility Accounting Method which assumes 65/35

debt-equity ratio, 5% interest on the outstanding debt, 48% income
tax rate and 7% return on the utility rate base, the twenty-year
average gas selling price is 62¢/10^6 Btu. With a 7.5% interest
rate and 9% return, the gas selling price would be about 65¢/MM
Btu.

It is therefore concluded that the Kellogg Coal Gasification
Process is an economically attractive process for the production
of substitute natural gas. Operation of the gasifier at 1200 psia
maximizes the direct production of methane thereby reducing the
heat supply requirement (and oxygen requirement) and improving
the economics. A comparison has shown that the heat requirement
in the gasifier is reduced by 44% at 1200 psia vs. 400 psia. Also,
over 99% of the carbon in the coal is converted to synthesis gas
owing to the strong catalytic action of the salt on the steam-carbon
reaction and the coal combustion reaction, as well as on the coal
hydrogenation reaction. For the above reasons, it is felt that
further development of this process as an alternate substitute
natural gas process or as a synthesis gas or hydrogen producer for
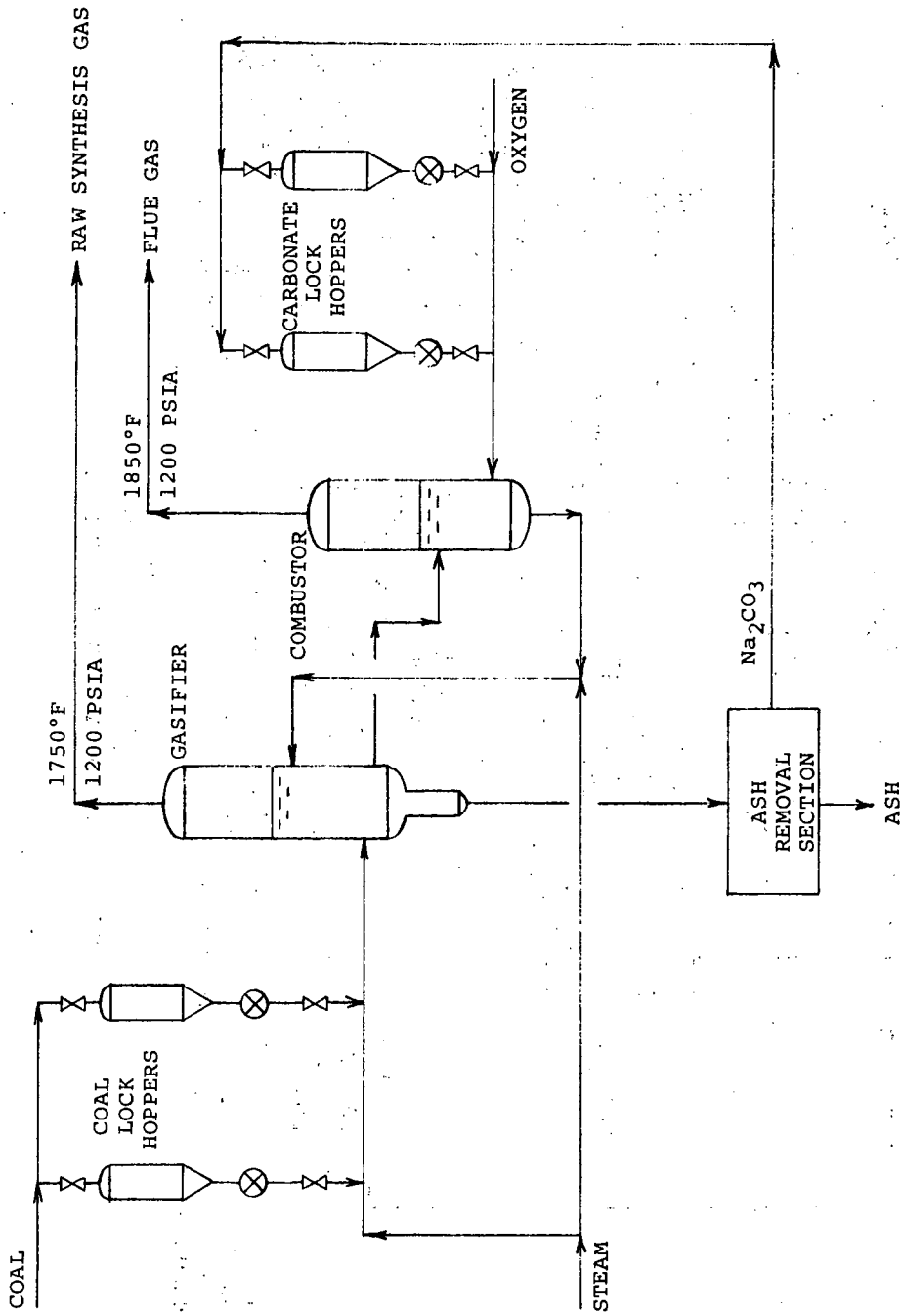hydrogasification or other uses is warranted.
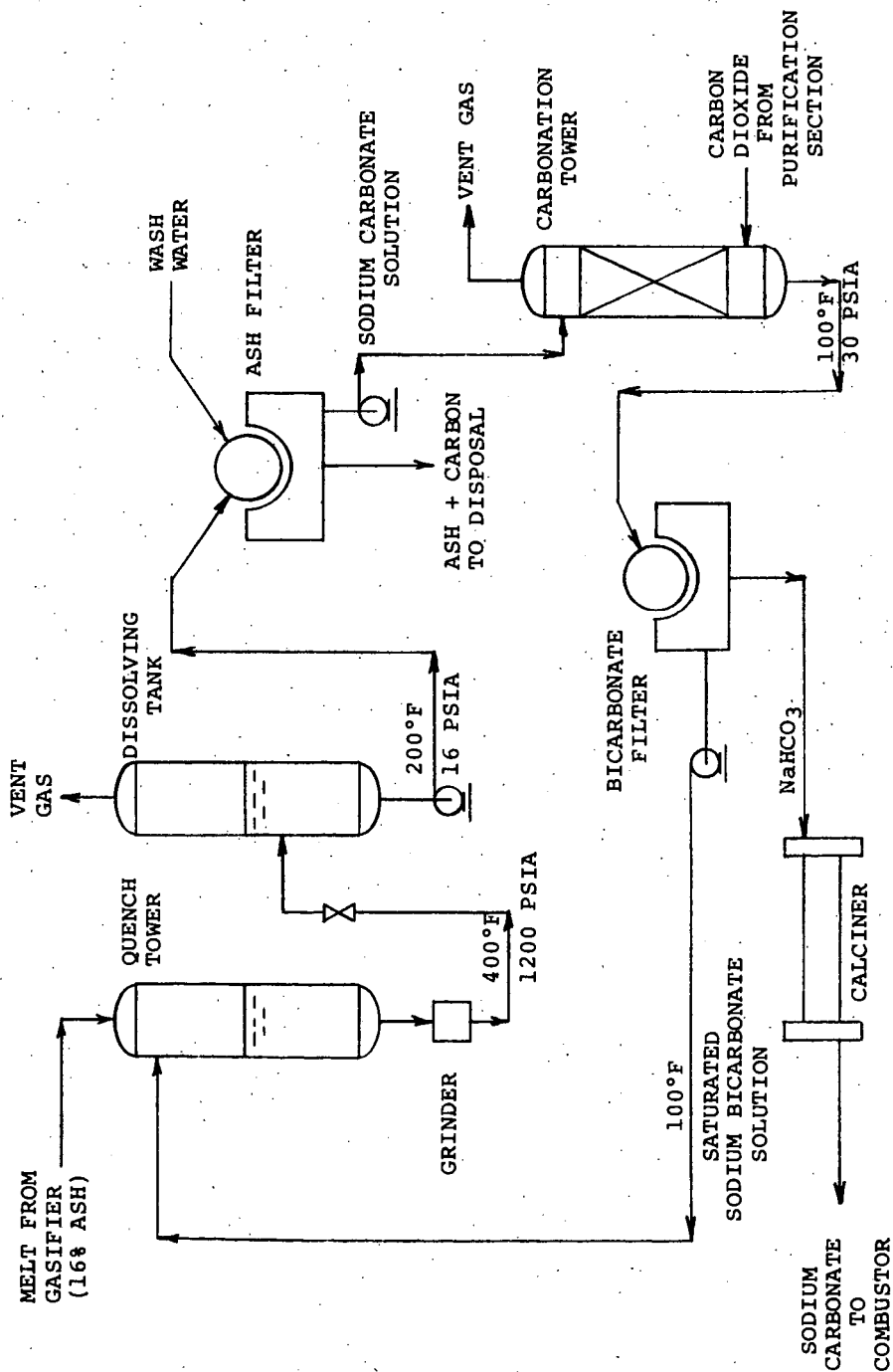
FIGURE 1

FLOW DIAGRAM FOR GASIFICATION SECTION

FIGURE 2

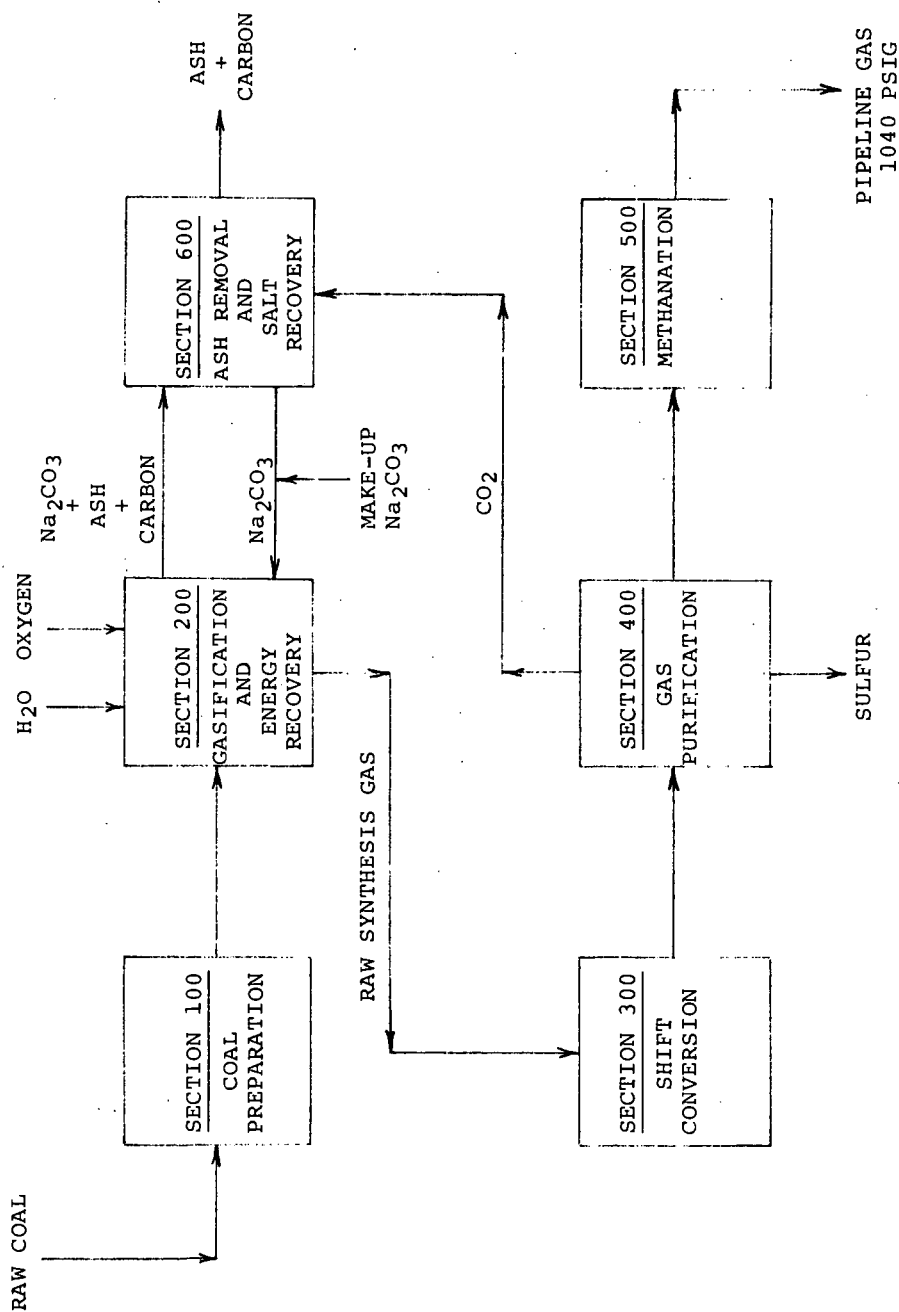FLOW DIAGRAM FOR ASH REMOVAL SECTION

FIGURE 3

BLOCK FLOW DIAGRAM

PIPELINE GAS FROM BITUMINOUS COAL

TABLE 1

INVESTMENT SUMMARY

PIPELINE GAS FROM BITUMINOUS COAL

KELLOGG MOLTEN SALT PROCESS

Basis: 262,000,000 SCFD of Pipeline Gas
        $(250 \times 10^9$ Btu/day)

Section	Title	INVESTMENT
100	Coal Storage and Preparation	$ 5,100,000
200	Gasification	39,100,000
	Oxygen Plant	30,600,000
300	Shift Conversion	10,000,000
400	Gas Purification	22,100,000
500	Methane Synthesis	4,300,000
600	Ash Removal	13,200,000
1100	Offsite Facilities	18,000,000
	TOTAL FIXED INVESTMENT	$142,400,000
	Working Capital	6,800,000
	TOTAL CAPITAL INVESTMENT	$149,200,000

TABLE 2

ESTIMATED GAS SELLING PRICE

PIPELINE GAS FROM BITUMINOUS COAL

KELLOGG MOLTEN SALT PROCESS

Basis: 262,000,000 SCFD of Pipeline Gas
(250 x 10^9 Btu/day)

Item	Gas Cost ¢/10^6 Btu
Bituminous Coal at $4/ton	17.6
Sodium Carbonate Make-up	2.3
Catalysts and Chemicals	1.0
Sulfur Credit at $10/ton	- 0.4
Power at 6 mills per kwh	6.7
Operating Labor	1.1
Maintenance & Supplies	5.6
Overhead & Supervision	3.8
Depreciation at 5% of Investment	8.6
Local Taxes & Insurance at 3% of Investment	5.2
TOTAL GAS PRODUCTION COST	51.5
GAS SELLING PRICE	61.9

STATUS AND DESIGN CHARACTERISTICS OF
THE BCR/OCR BI-GAS PILOT PLANT

By

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I.   INTRODUCTION

Research and development was initiated in 1965 on the generation of
data required for the design of a pilot plant for the production of high-
Btu pipeline gas based on the concept of two-stage super-pressure entrained
gasification of coal. Experimental work on the program has progressed
successively through three levels of scale-up and has now reached the point
where operation of a fully integrated coal-to-pipeline gas pilot plant is
considered to be the next logical step in the development of the process.

Initial experiments were conducted in rocking autoclaves using 5-
gram charges of coal. Resultant data confirmed the basic assumption that
a high yield of methane could be obtained directly from coal by reaction
with steam at elevated temperatures and pressures.[1]*** Flow experiments
in an externally-heated reactor confirmed the assumption that higher meth-
ane yields could be obtained from coal entrained in steam-synthesis gas
mixtures at short residence times.[1] Recent work in an internally-
fired 100 lb/hr flow system has further confirmed the basic principle
and advantages of the two-stage super-pressure concept, and has provided
design data for the integral two-stage gasifier.[2]

This paper presents the design characteristics of a fully integrated
coal-to-pipeline gas pilot plant based on a 5 ton/hr two-stage, super-
pressure, entrained oxygen-blown gasifier, otherwise designated as the
BCR/OCR BI-GAS PROCESS.

II.   BASIC PRINCIPLES AND ADVANTAGES OF
THE BI-GAS PROCESS

A.   Basic Principle of BI-GAS Process

In the two-stage super-pressure gasification process[3], fresh
pulverized coal is introduced into the upper section (Stage 2) of the

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*** Numbers in parenthesis indicate references listed under
    "Literature Cited."

gasifier at pressures in the range of 70 to 100 atm. (See Figure 1.) Here, the coal comes in contact with a rising stream of hot synthesis gas produced in the lower section (Stage 1) and is partially converted into methane and more synthesis gas.

The residual char entrained in the raw product gas is swept upward and out of the gasifier. The char is separated from the product gas stream and recycled to the lower section (Stage 1) of the gasifier.

In the lower section, the char is completely gasified under slagging conditions with oxygen and steam, producing both the synthesis gas and the heat required in the upper section (Stage 2) for the partial gasi-fication of the fresh coal.

B. Status of Development

The research and development to date has been directed primarily toward development of the technology of Stage 2 and optimization of operating conditions for production of a gas suitable for conversion to high-Btu pipeline gas by catalytic methanation. Through laboratory-scale studies and process and equipment development, design data for Stage 2 is now available. The technology of Stage 1, while not complete, has been well advanced by others and is considered adequate for design of a pilot-scale unit. Construction and operation of an integral two-stage gasifier is now considered the next step in development of the BI-GAS process.

C. Advantages of Two-stage Gasification

When the two-stage gasifier is operated on oxygen at system pressures of 1,000 psi or higher, the resultant raw gas is an excellent feedstock for upgrading to substitute natural gas.

When the gasifier is operated on air at moderate system pressures, a gas is produced which may be readily desulfurized and cleaned to yield a pollution-free fuel gas with a heating value of about 175 Btu/scf.

The two-stage, oxygen-blown, super-pressure gasifier offers several advantages in the production of substitute natural gas. A high yield of methane is obtained directly from coal and subsequent processing of the output gas is minimized. Because it is an entrained rather than a fixed or fluidized-bed system, all types of coal should be amenable without prior treatment for use in this gasifier. All the feed coal is con-sumed in the process; principal by-products are slag for disposal and sulfur for sale. The two-stage gasifier, being an integral unit, is relatively simple in design and amenable to scale-up to most any size. Also, the conditions in Stage 2 are such that no tar and oils are formed in the gasification process.

III. DESCRIPTION OF BI-GAS PILOT PLANT

For the past two years, planning has been underway for the further development of two-stage gasification. The plans include a much broader concept than the erection and operation of a pilot-scale gasifier. Inasmuch as the equipment, personnel, and other facilities necessary for the pilot testing of the BCR two-stage process will be usable in the solution of problems related to other gasification processes under development, it appears prudent that a broader view should be taken in planning of pilot-plant research.

Further stimulating this concept is the growing interest in the development of a new approach to power generation incorporating coal gasification at the power station; this approach would use an air-blown rather than an oxygen-blown gasifier, followed by large high-temperature gas turbines.

Early discussions with the Office of Coal Research resulted in the concept of a multipurpose research integrated facility, and a technical evaluation and cost estimate of such an integrated research facility was made with the assistance of Koppers Company, Inc.(4)

The research facility as currently planned will ultimately embody alternate systems for each of the major unit operations in the conversion of run-of-mine coal, or char, to high-Btu pipeline gas; to sulfur-free, low-Btu fuel gas; to industrial gases; and to MHD fuel gas. Three types of gasification units will be provided: an air-blown, medium-pressure, two-stage gasifier; an oxygen-blown, super-pressure, two-stage gasifier; and a low-pressure, multi-stage, fluidized-bed gasifier.

The initial pilot plant is now being designed for the multipurpose research pilot plant facility (MPRF); it is based on the oxygen-blown version of the two-stage gasifier and will include all facilities needed for converting coal into high-Btu pipeline-quality gas.(5) In addition to the basic gasifier, the system would include facilities for: coal crushing, sizing, and drying; high-pressure coal feeding system; dust and heat recovery; CO-shift; acid gas removal; and methanation. (See Figure 2.) Space will be provided for future installation of special beneficiation equipment for preparation of enriched gasifier feedstock.

A. Coal Beneficiation, Storage, and Handling

Initial operations of the 5 ton/hr pilot plant will be confined to the testing of washed coals. Prepared coals, with an approximate size consist of 1-1/2 inch by 0, will be delivered to the plant. However, the plant layout will provide for the future addition of coal beneficiation facilities to permit the receiving and processing of run-of-mine coals. Figure 3 is a schematic diagram of these facilities.

The washed coals for the early test work will be received via truck hopper and transferred to two 450-ton storage bins by a conveyor system, equipped with belt scales, tramp iron magnet, and automatic sampler. Similarly, run-of-mine coal may be unloaded at the truck hopper, conveyed to a primary crusher and thence to storage, or crushed coal may be delivered directly to the final crushing and screening station before the beneficiation plant.

Laboratory studies have indicated that by proper choice and beneficiation of the gasifier feedstock, increased yields of methane may be expected in Stage 2 of the gasifier. These studies, aimed at concentrating the gas-rich macerals, are being continued in an effort to develop systems more effective than those currently available. The proposed initial coal beneficiation system shown schematically in Figure 4 will include the following circuits:

air flow tables for treating 3/4 inch x 0, 38 inch x 0, or 1/4 inch x 0 coals;

Deister tables for treating 3/8 inch x 28M or 1/4 inch x 28M coals;

heavy media cyclones for treating 3/4 inch x 28M coal or 1/2 inch x 28M and,

flotation cells for treating the minus 28 mesh coal.

The coals to be tested in the 5 ton/hr pilot plant will range in rank from lignite to high volatile bituminous. The tentative program includes the testing of: Pittsburgh Seam; Illinois No. 6 Seam, or West Kentucky No. 11 Seam; Elkol Seam from Wyoming; and, North Dakota Lignite. In addition, it is planned to conduct tests on chars produced from other coal conversion processes now under development.

The system for handling and preparing the washed coal for feed to the gasifier is shown in Figure 5. The coal is delivered from the 450-ton storage bins by means of weigh feeders and conveyor belt to a combination dryer and pulverizer equipped with a classifier for returning oversize coal to the pulverizer and with primary and secondary separators for receiving the pulverized coal. The dry coal, pulverized to approximately 70 percent minus 200 mesh, is transferred from these receivers by screw conveyor to a pulverized coal storage bin.

The system also provides for the handling of flux materials. The flux may be mixed with the coal and fed to Stage 2 of the gasifier or the flux may be fed separately to Stage 1.

B.   Coal Feeding

A major mechanical problem associated with processes designed for the total gasification of solid fuels in suspension is the injection of the pulverized fuel into the systems at a uniform feed rate at pressures of 50 to 100 atmospheres. Since the conception of high pressure oxygen gasification of coal, with its apparent advantages over atmospheric pressure operation for the production of pipeline gas, considerable effort has been devoted to developing feeding systems for high-pressure operation. Feed systems that have been, or currently are, under development include fluidized feeders, lock-hopper systems, slurry systems, high-pressure jet feeders, and various designs of table, rotary, and screw feeders.

The slurry feed system would appear to provide the least complicated means of transport and of obtaining a uniform feed rate providing the ratio of solids to liquid is constant. The use of water as the vehicle introduces a thermal disadvantage and creates a problem in that the amount of water required for a pumpable slurry is in excess of the water required for the steam reactant. Attempts to flash off the excess steam just prior to entry to the gasifier reactant nozzles have met with little success to date. The use of oil as the vehicle would eliminate these problems associated with the water slurry and, of course, would result in increased gas production.

To overcome the disadvantages of the lock-hopper and slurry systems, BCR conceived a design of a piston type feeder which was confirmed by laboratory experiments. A nominal 600 lb/hr prototype unit incorporating certain modifications was purchased for test operation.

As shown in Figure 6, the piston coal feeder has two main operating sections, a low-pressure section and a high-pressure section. In the low pressure section, pulverized coal is transferred pneumatically from a storage tank to the weigh tank, F-1, using an inert gas as the transport medium. From the weigh tank, F-1, at atmospheric pressure, the coal flows by gravity through a pneumatically operated ball valve into a low pressure injector vessel, F-2. The injector vessel, F-2, is pressurized with recycle product gas to about 90 psi.

In the high pressure section, while the piston is in the raised position, the coal-gas mixture in the F-2 vessel is vented into the high pressure piston injector vessel, F-3. The ball valve between the F-2 and F-3 vessels is then closed and the F-3 vessel is pressurized to about 100 lb above that of a high-pressure receiver, F-4. The valve between the F-3 vessel and the F-4 vessel is then opened; by so doing, the coal plus gas is allowed to flow into the receiver vessel. Hydraulic pressure lowers the specially designed piston and completes the transfer of the residual coal-gas charge into the high-pressure receiver. The valve between the two vessels is closed, the small quantity of high pressure gas remaining in the injector vessel is vented to a gas holder, and the piston is returned to its original position by means of a hydraulically operated lift rod. The entire cycle is then repeated automatically.

In test operations at the BCR laboratories, performance data have been collected on the gas consumption, capacity, and reliability of the system. In these tests, the unit was operated routinely, injecting coal at rates up to 800 lb/hr into a receiving vessel pressurized to 1,400 psi.

For the 5 ton/hr pilot plant, it is planned to install two piston feeders, each with a capacity of 2.5 ton/hr. In addition, the layout for the pilot plant will provide for the installation of an alternate lock-hopper system to deliver coal to the high-pressure coal feed tanks represented as F-4 in Figure 6. Thus, in the event extended development work on the piston feeder is required, operation of the gasification plant can be sustained by means of the lock hopper-feed arrangement.

Various designs of feeders for delivering the coal from the high-pressure receivers to the transport lines to the gasifier are being studied. Tentatively, it is planned to use a specially designed screw feeder for this application, but further development work and testing of feeders is required to establish the most suitable feeder for this purpose. Process gas will be used for the transport medium and, at pressures of about 100 atm, the weight ratio of coal to gas will be about 8:1.

C.   Coal Gasification

As stated in the general description of the pilot plant, the gasifier will be an oxygen-blown, two-stage unit designed for coal feed rates up to 5 ton/hr and a maximum operating pressure of 1,500 psi. The unit will be adaptable to all ranks of coals.

The outside diameter of the gasifier is 5.5 feet, and the diameter inside the refractories is 3 feet. The heights of Stages 1 and 2 are 6 feet and 14 feet, respectively, and the height of the slag quench and removal section is 13 feet. Velocities in each stage will vary with changes in feed rate and operating pressure. The expected minimum gas velocities for Stages 1 and 2 are 0.27 ft/sec and 0.15 ft/sec, respectively; corresponding maximum residence times are 24 and 73 seconds. The expected maximum velocities are 2.3 and 1.12 ft/sec for Stages 1 and 2, respectively, and the corresponding minimum residence times are 2.9 and 9.8 seconds. The gas exit temperature will be about 1700 F and, as shown in Table 1, the composition of the gas will be approximately 16 percent carbon dioxide, 28 percent hydrogen, and 7 percent methane, with the balance being water and less than 1 percent each of nitrogen and hydrogen sulfide.

A simplified process flow diagram with major flow rates and operating conditions for a coal feed rate of 5 ton/hr is shown in Figure 7. A material balance is given in Table 2.

Dry, pulverized coal, conveyed with steam, is fed into the upper stage (Stage 2). The coal is devolatilized and partially gasified in Stage 2 by the steam and the hot gases and char rising from the lower reaction zone (Stage 1).

TABLE 1. GAS STREAM COMPOSITIONS FOR OXYGEN-BLOWN TWO-STAGE SYSTEM
(Volume Percent)

Component Gas	1 Gasifier Product	2 CO-shift Feed	3 Acid Gas Removal Plant Feed	4 Acid Gas Removal Plant Product	5 Final Pipeline Gas
CO_2	16.25	12.52	31.06	0.23	0.63
CO	23.60	18.19	14.63	21.37	0.11
H_2	28.41	21.90	45.00	65.72	4.79
CH_4	7.04	5.43	8.07	11.79	92.36
N_2	0.45	0.35	0.52	0.76	2.11
H_2S	0.55	0.43	0.63	0.00	0.00
H_2O	23.70	41.18	0.09	0.13	0.00
Total	100.00	100.00	100.00	100.00	100.00

TABLE 2. MATERIAL BALANCE FOR 5 TON/HR
OXYGEN-BLOWN TWO-STAGE GASIFIER

Input	Weight, lb/hr	
Stage 2		
Coal	10,000	
Transport Gas Recycle	(1,919)	
Steam	8,774	18,774
Stage 1		
Char Recycle	(9,408)	
Steam Feed Gas Recycle	(275)	
Steam	2,880	
Oxygen (99.5%)	6,361	9,241
Total Input		28,015
Output		
Stage 2		
Raw Product Gas	27,331	
Recycle Gas	(2,194)	
Recycle Char	(9,408)	27,331
Stage 1		
Slag	684	684
Total Output		28,015

The products leaving the gasifier are first cooled; the entrained char is then separated from the gas, collected, and reinjected with oxygen and additional steam into the Stage 1 reaction zone.

Molten slag formed in Stage 1 drops through a water spray into a reservoir of liquid water below. The resulting granulated slag particles are periodically removed through a lock hopper system.

D. Waste Heat Recovery and Char Recycle System

Details of the waste heat recovery and char recycle system are shown in Figure 8. By means of a heat exchanger, essentially a waste heat boiler, and direct water sprays, the temperature of the char and gas will be decreased to about 900 F. A multi-cyclone unit will be used for separating the char from the gas. By sequential operation of lock hoppers, the char is dropped by gravity into the gasifier char feed bin.

Similar to the situation with coal feeders, various types of char feeders are being investigated. Present plans for feeding hot char to Stage 1 are to use an arrangement of a rotary valve at the base of the feed bin to deliver char into a standpipe which in turn will feed a specially designed screw feeder. Maintaining a uniform head of char with uniform bulk density on the screw feeder facilitates the problem of maintaining uniform feed rates. The gasifier will be equipped with three reactant or char burning nozzles. Each nozzle will be equipped with a separate feed system.

About 95 percent of the char will be returned by the multi-cyclone. Approximately two-thirds of the char entering Stage 1 is gasified and about one-third is recycled. Thus, about 1.6 lb of fixed carbon is recycled to Stage 1 for each pound of fixed carbon entering with the coal in Stage 2.

The gas, with finely dispersed char, passes from the multi-cyclone to a second heat exchanger, steam generator, where the gas temperature is reduced to 650 F in preparation for the carbon monoxide shift reaction. After the heat exchanger, the gas is filtered to remove the fine char. Currently, a sand bed filter is under investigation for the removal of fine char. If successful, it will eliminate the need for further cooling of the gas and subsequent reheating for the CO shift. If solid bed filters are unsatisfactory, it may be necessary to resort to bag-type filters which presently require gas temperatures below 500 F, or to wet scrubbing which will lower the gas temperature even further. In either case it would be necessary to reheat the gas prior to the CO shift.

The char recycle system will provide for either returning the fine char to the gasifier char feed bin or for removing it from the system.

E. Carbon Monoxide Shift

The purpose of the carbon monoxide shift conversion is to adjust the hydrogen to carbon monoxide ratio in the gas to 3.1 to 1 as required for the methane synthesis. As shown in the simplified process flow diagram, Figure 9, the facilities will include catalyst guard filters, reactors, and the appropriate heat exchangers.

Gas from the heat recovery and char removal system will be fed to the CO shift facility at a temperature of 650 F and a pressure of about 1000 psi. In the catalyst guard filters, any olefins in the gas will be hydrogenated; sulfur compounds such as carbonyl sulfide will be converted to hydrogen sulfide; and any fine char remaining in the gas will be trapped. By providing two units, the catalyst in the off-stream unit may be regenerated with steam and air.

Superheated steam will be added to the gas entering the reactors to adjust the steam to dry gas ratio to 0.7. Composition of the feed gas to the reactors is shown in Table 1. The extent of reaction and thus the hydrogen to carbon monoxide ratio of the exit gas stream will be controlled by appropriate settings of the following operating conditions: (1) the steam/gas ratio, (2) the amount of gas bypassed, and (3) reactor inlet gas temperature.

For the 5 ton/hr pilot plant it is expected that the bypass stream will be about 40 percent of the feed gas and that 95 percent of the carbon monoxide in the remaining 60 percent will be converted to hydrogen and carbon dioxide.

The exothermic heat release from the reaction will raise the gas temperature from 650 F to about 865 F. After mixing the converted gas with the bypass stream, the gas is cooled to 95 F. The composition of the mixed gas stream, having a H_2/CO ratio of 3.1, is shown in Table 1.

F. Acid Gas Removal

From a study of some 35 available acid gas removal processes, it was concluded that the criteria established for the BI-GAS process were best satisfied by those processes employing an organic solvent as the stripping medium and which selectively remove the hydrogen sulfide and carbon dioxide. Such processes include the Purisol, the Rectisol, and the Selexol.

Figure 10 is a simplified process flow diagram of the acid gas removal and sulfur recovery based on the Purisol process and a Claus unit for recovery of elemental sulfur.

The gas from the CO shift converter will first pass through an absorber where the hydrogen sulfide will be absorbed in N-methyl-2-pyrrolidone. The hydrogen sulfide will be steam stripped from the solvent in a separate tower and fed to the Claus sulfur recovery plant. The solvent will be recycled. The process gas from the hydrogen sulfide absorber will pass through a second absorber where the carbon dioxide is absorbed. The carbon dioxide will be stripped from the solvent in a separate vessel and vented to atmosphere; the solvent is recycled.

To satisfy pipeline gas specifications the hydrogen sulfide content of the gas must be below 0.25 grain per 100 scf. The composition of the gas leaving the acid gas removal section is shown in Table 1.

G. Methanation

The most advanced designs of methanation processes - that is, the conversion of carbon monoxide and hydrogen to methane, are based on fixed-bed catalytic reactors. However, in the interest of furthering research in this area, current laboratory work is directed toward development of a methanation system based on a fluidized-bed catalytic reactor.

A simplified process flow diagram of the methanation step based on using a fluid-bed reactor is shown in Figure 11.

The methanation reaction is highly exothermic, and temperature control is critical. Below 500 F the carbon monoxide reacts with the nickel catalyst to form nickel carbonyl, resulting in loss of catalyst and the production of a hazardous gas. Above 500 F carbon formation occurs by the reaction of hydrogen with carbon to form water and elemental carbon, resulting in a decrease in yield.

The fluid-bed reactor should provide for better heat transfer than is possible with fixed-bed reactors. With sufficient cooling coils and heat exchange capacity, the exothermic heat should be controllable at higher levels of carbon monoxide concentration in the feed gas than is now possible.

The temperature of the feed gas will be in the range of 550 to 625 F. In the pilot plant, provisions will be made to introduce the feed gas at three levels to permit distribution of heat. (See Figure 11.) With the appropriate arrangement of cooling coils and heat exchangers, it is expected that process gas containing about 20 percent carbon monoxide can be fed directly to the methanator.

The final pipeline gas, having a gross calorific value of about 950 Btu/scf, will have a composition as shown in Table 1.

IV. GENERAL FACILITIES

The research facility now being planned, requiring approximately 25 acres, is to be located near U. S. Highway 119 just north of Homer City, Pennsylvania, not far from the BCR research center at Monroeville, Pa. The facility would be laid out to accommodate the proposed experimental systems plus general utilities. Space for expansion would also be provided, especially in the gas processing area.

Water for cooling purposes, steam generation, and fire protection will be supplied from Two Lick Creek which borders on the plant property. Utilization of this water will require installation of facilities for pumping, cooling, filtering, and chemical treatment. City water will be available for limited required uses.

In a commercial plant, steam requirements may be satisfied by use of heat exchange equipment in the gas treatment section of the plant. However, a steam boiler will be provided for the pilot plant to assure operation of the gasifier exclusive of the gas treatment facilities.

Adequate electrical power will be furnished to the plant so no generating equipment is required. The necessary substation and motor control rooms will be installed.

Buildings will be provided for office, laboratory, maintenance, and other services as required. Miscellaneous facilities will include roads, parking lots, communication system, etc. All effluents will be treated and all waste products collected to eliminate environmental pollution.

V. SUMMARY

The design characteristics of a fully integrated coal-to-pipeline gas pilot plant based on the BI-GAS process have been delineated. At the present time, these are being incorporated in an engineering bid package scheduled for completion in October, 1971. With appropriate funding, procurement and erection of a 5 ton/hr pilot plant on the site at Homer City, Pennsylvania, may be reasonably expected to be completed by early 1974.

ACKNOWLEDGMENT

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2. Grace, R. J., Glenn, R. A., and Zahradnik, R. L., "Gasification of lignite by the BCR two-stage super-pressure process," AIChE, Symp. Synthetic Hydrocarbon Fuels from Western Coals, Denver, Colorado, 1970.

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4. Glenn, R. A., "A multipurpose research pilot plant facility featuring BCR two-stage super-pressure coal gasification," ACS Div. Fuel Chem., Chicago, Ill., September, 1970.

5. Glenn, R. A., "Status of the BCR two-stage super-pressure process," Am. Gas Assoc., Third Synthetic Pipeline Gas Symposium, Chicago, Ill., 1970.
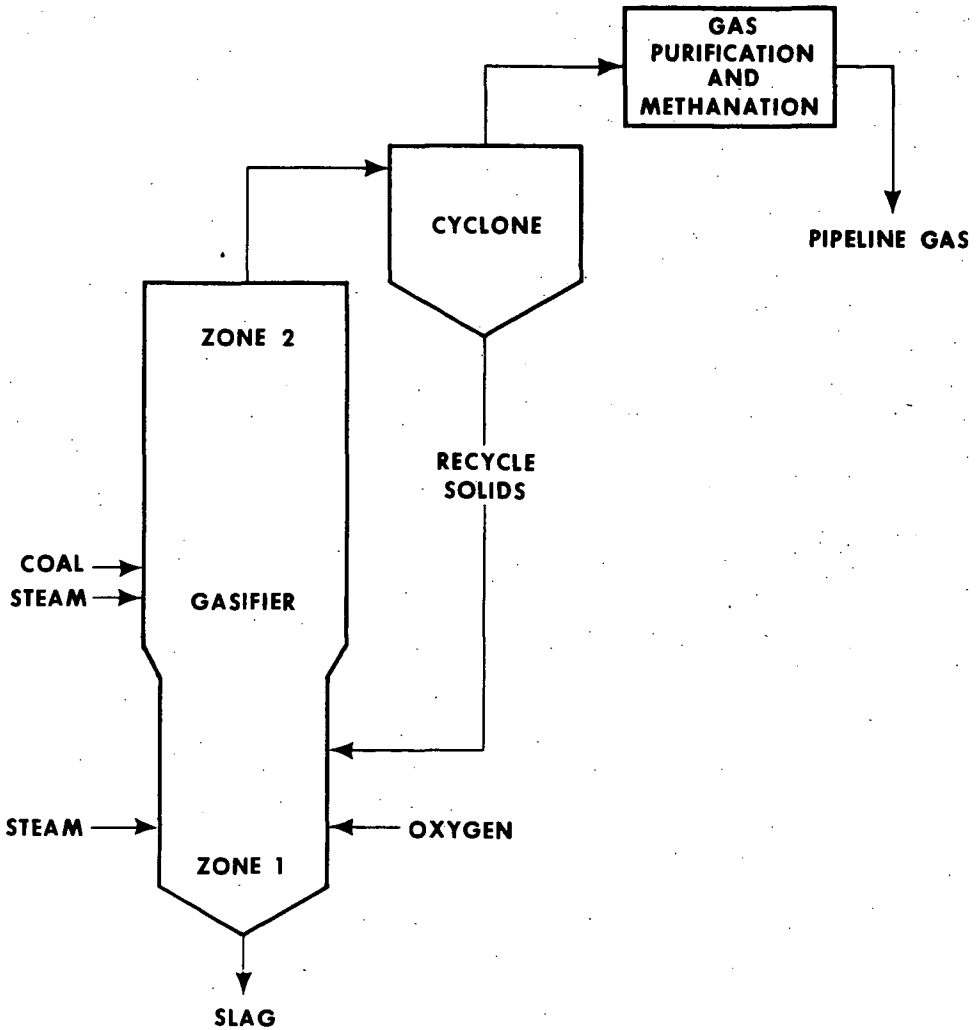
Bituminous Coal Research, Inc. 8022G17

Figure 1. Simplified Flow Diagram for Two-stage
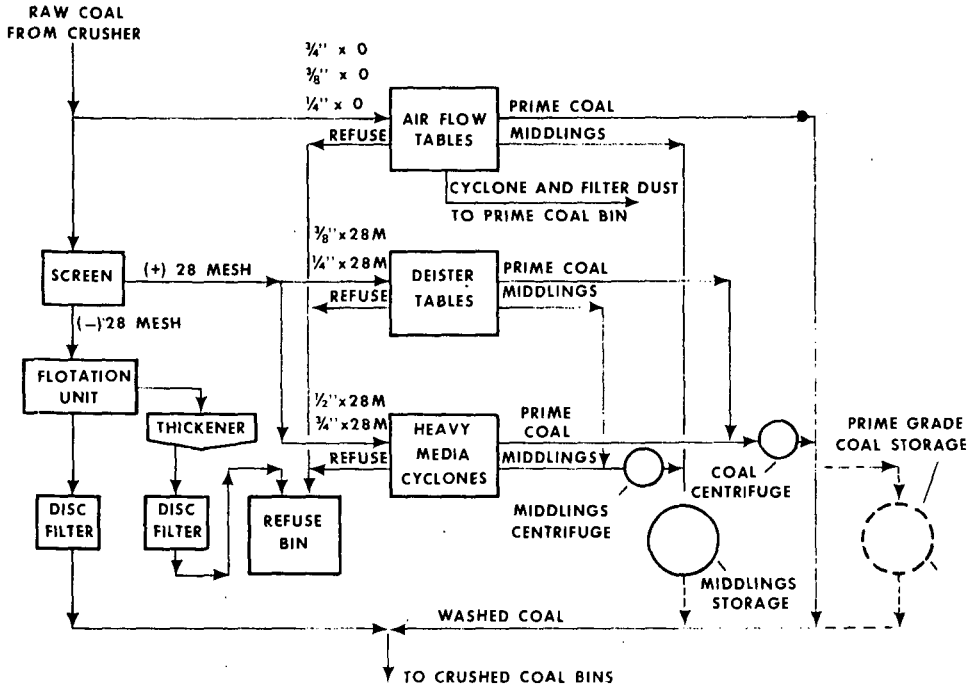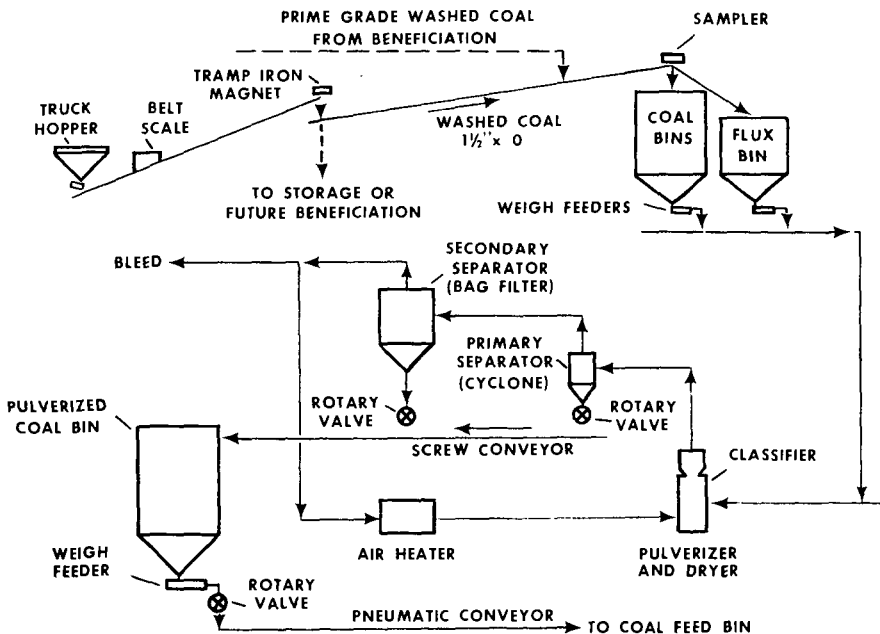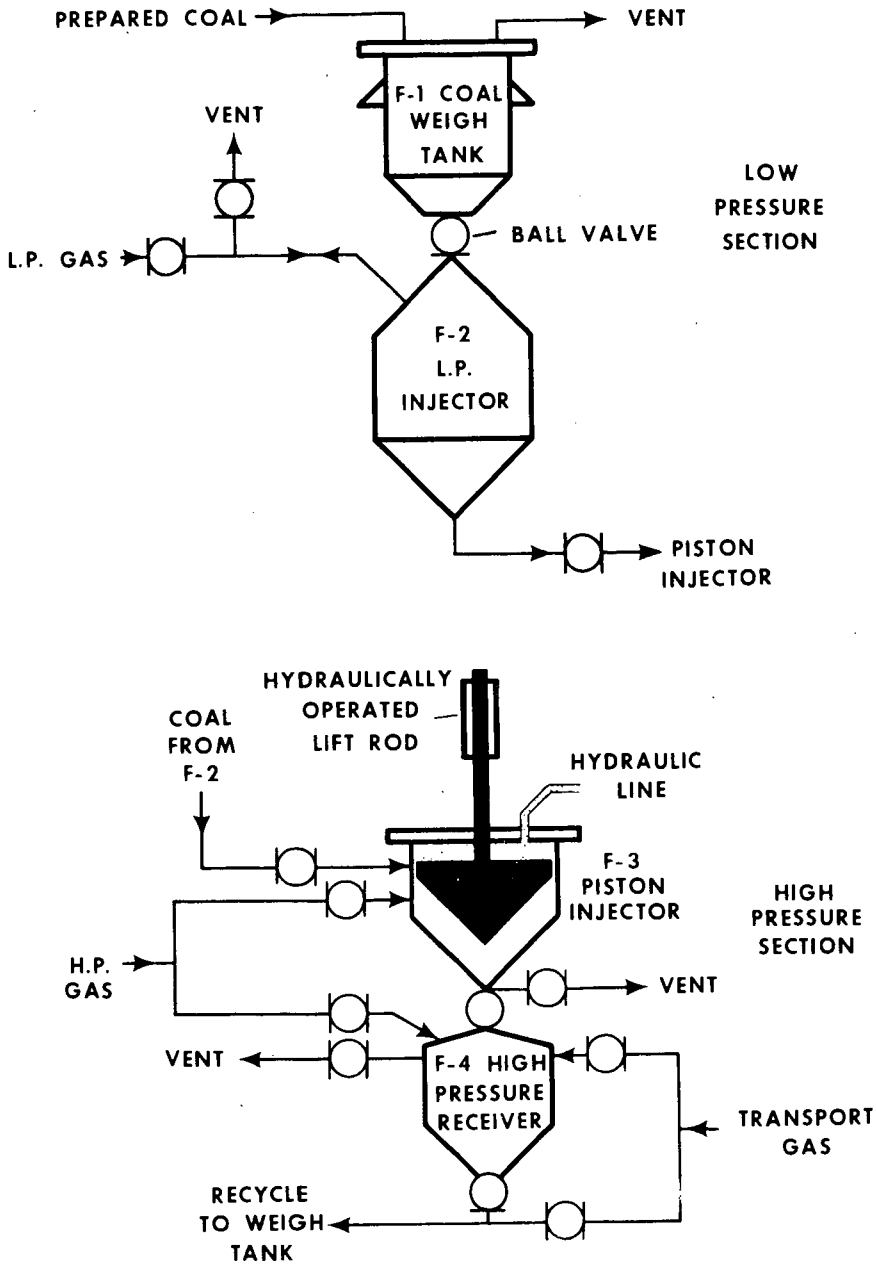Super-pressure Gasifier

Bituminous Coal Research, Inc. 8022G18

Figure 2.   BCR/OCR BI-GAS Pilot Plant Flow Diagram

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Figure 3.   Coal Handling and Storage System Flow Diagram

RAW COAL
FROM CRUSHER

¾" x 0
⅜" x 0
¼" x 0

AIR FLOW TABLES

PRIME COAL
MIDDLINGS
REFUSE

CYCLONE AND FILTER DUST
TO PRIME COAL BIN

SCREEN

(+) 28 MESH

⅜"x28M
¼"x28M

DEISTER TABLES

PRIME COAL
MIDDLINGS
REFUSE

(−) 28 MESH

FLOTATION UNIT

THICKENER

½"x28M
¾"x28M

HEAVY MEDIA CYCLONES

PRIME COAL
MIDDLINGS
REFUSE

DISC FILTER

DISC FILTER

REFUSE BIN

MIDDLINGS CENTRIFUGE

COAL CENTRIFUGE

PRIME GRADE COAL STORAGE

MIDDLINGS STORAGE

WASHED COAL

TO CRUSHED COAL BINS

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Figure 4. Coal Beneficiation Flow Diagram

PRIME GRADE WASHED COAL
FROM BENEFICIATION

SAMPLER

TRUCK HOPPER

BELT SCALE

TRAMP IRON MAGNET

WASHED COAL
1½"x 0

COAL BINS

FLUX BIN

TO STORAGE OR
FUTURE BENEFICIATION

WEIGH FEEDERS

BLEED

SECONDARY SEPARATOR
(BAG FILTER)

PRIMARY SEPARATOR
(CYCLONE)

PULVERIZED COAL BIN

ROTARY VALVE

ROTARY VALVE

SCREW CONVEYOR

CLASSIFIER

WEIGH FEEDER

ROTARY VALVE

AIR HEATER

PULVERIZER AND DRYER

PNEUMATIC CONVEYOR

TO COAL FEED BIN

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Figure 5. Coal Handling and Preparation System Flow Diagram

PREPARED COAL ⟶   ⟶ VENT

VENT

L.P. GAS ▶

F-1 COAL WEIGH TANK

BALL VALVE

LOW PRESSURE SECTION

F-2 L.P. INJECTOR

PISTON INJECTOR

COAL FROM F-2

HYDRAULICALLY OPERATED LIFT ROD

HYDRAULIC LINE

F-3 PISTON INJECTOR

HIGH PRESSURE SECTION
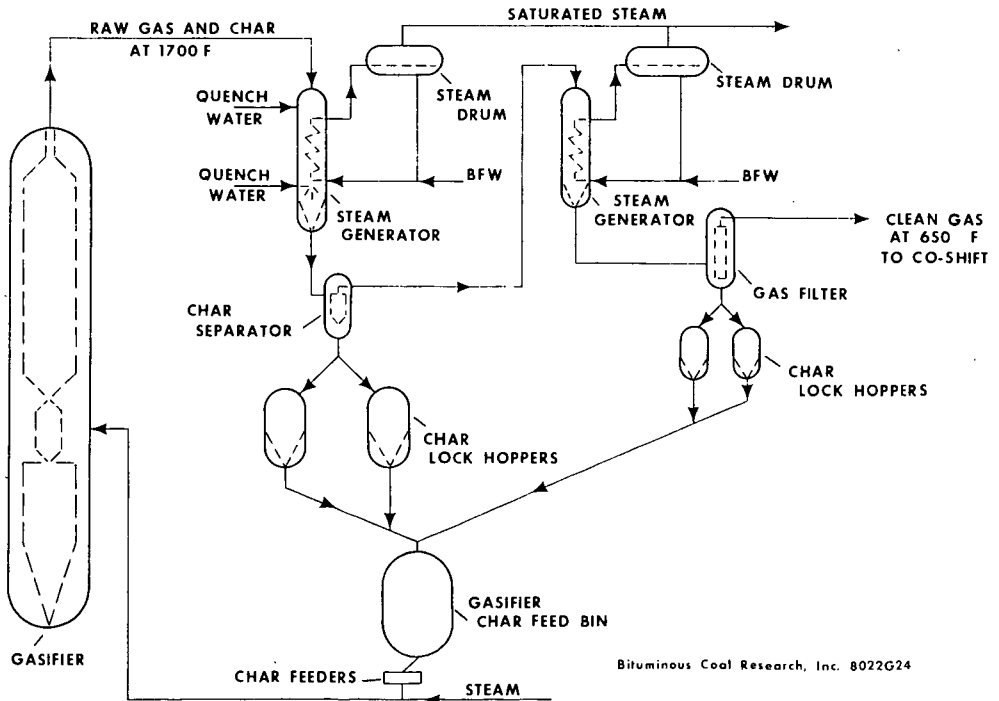
H.P. GAS

VENT

VENT ◀

F-4 HIGH PRESSURE RECEIVER

TRANSPORT GAS

RECYCLE TO WEIGH TANK ◀

Bituminous Coal Research, Inc. 8022G22

Figure 6. Piston Feeder Diagram
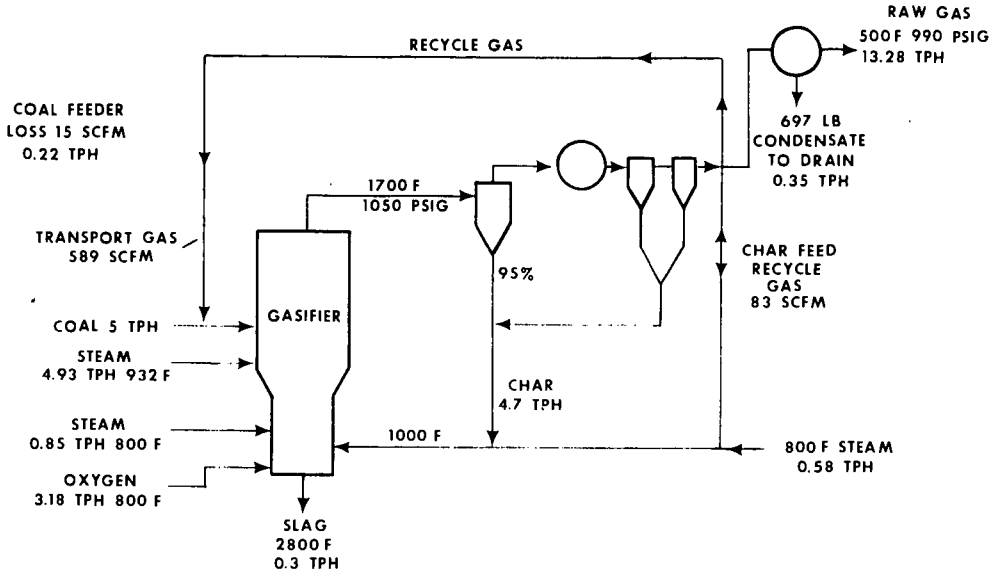
Figure 7. Process Flow Diagram for 5 TPH Oxygen-blown Two-stage Gasifier

Figure 8. Waste Heat Recovery and Char Recycle Flow Diagram
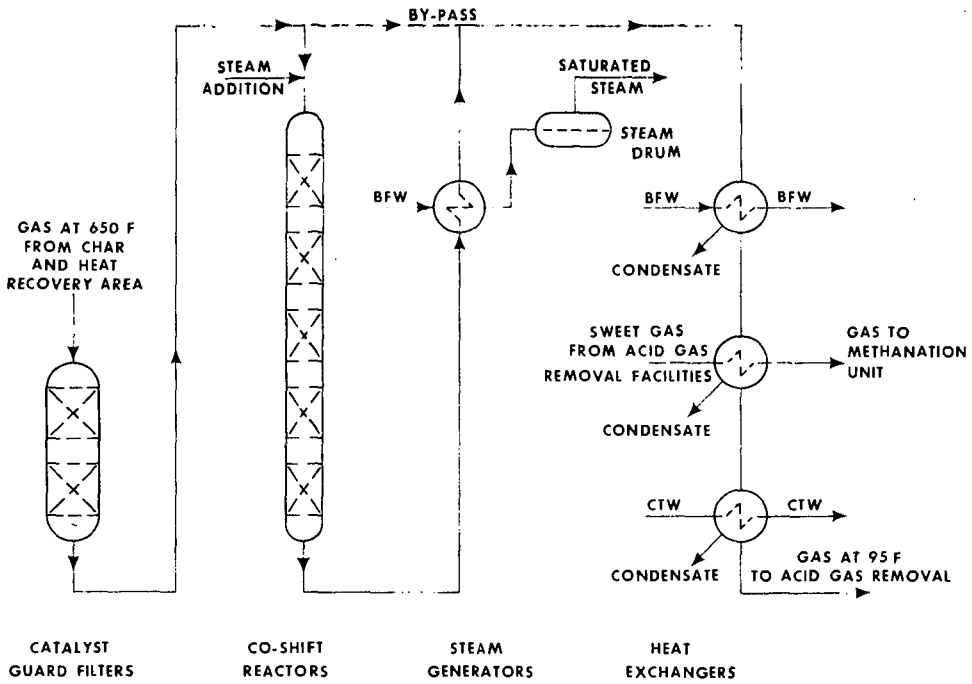
Figure 9. CO-shift Flow Diagram
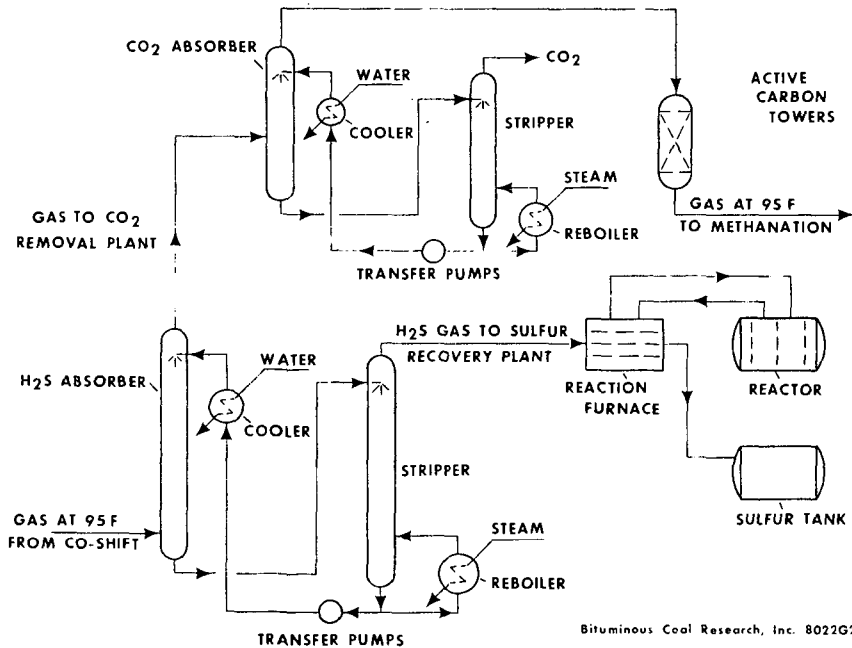
Figure 10. Acid-gas Removal and Sulfur Recovery Flow Diagram
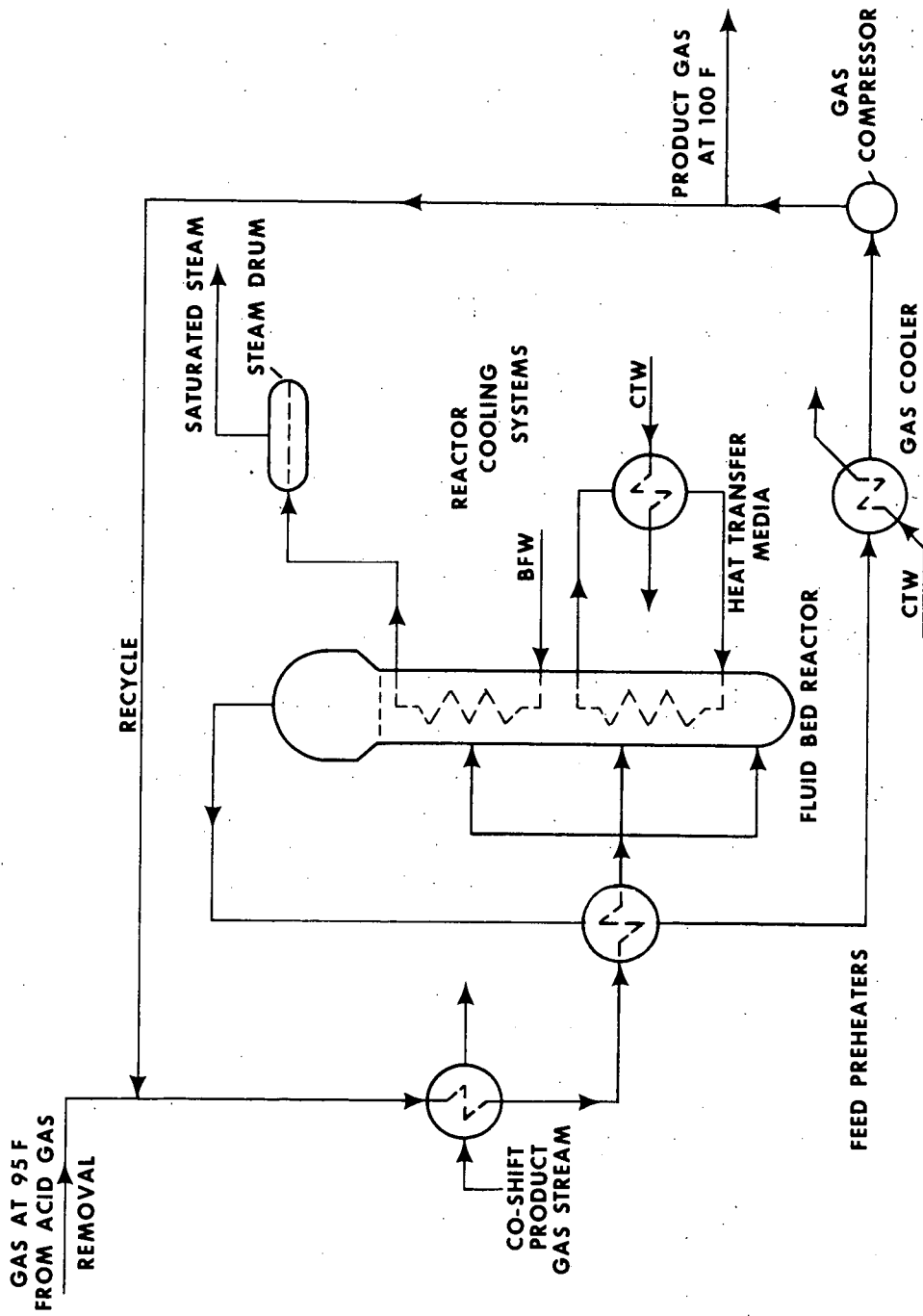
Figure 11. Methanation Flow Diagram

THE SYNTHANE COAL-TO-GAS PROCESS: A PROGRESS REPORT

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INTRODUCTION

National energy needs demand the development of a supplemental natural gas from coal. This is of deep concern to the pipeline industry, the gas industry, and the coal industry. What is needed is an operable economic process. The Bureau of Mines at Bruceton, Pa., has developed a process for making a high-Btu gas from coal--the Synthane Process. The steps of gasification, gas purification, and methanation have been tested to the point that a prototype plant is being designed.

THE OVERALL PROCESS

The Synthane Process is shown schematically in figure 1. The main components are the gasifier, the shift converter, the purification system, and the catalytic methanator. The water-gas shift step is commercial, and no work has been done on this step. The Bureau hot carbonate purification work was developed about 10 years ago [1] and has been adopted commercially but is no longer being researched. The gasifier and methanator are being tested at the present time.

The purified gas has a heating value of about 500 Btu/cu ft before methanation and a heating value exceeding 900 Btu/cu ft after methanation.

The Gasifier

The gasifier is shown in figure 2. It is a fluid-bed type, operating at 40 atmospheres (600 psia) and up to 1,000° C (1,830° F). The coal, 70% through 200 mesh, is dropped through the pretreater with oxygen and steam (or CO_2) at 400° C (750° F), where it is rendered non-caking. The decaked coal then falls into the carbonization zone and finally to the gasification zone where it is gasified with steam plus oxygen.

Results of the latest tests with free-fall pretreatment are shown in table 1, where the data are compared with assumptions made for the economic study. The data shown have met or exceeded all assumptions made, considering the limitations of our 6-foot-high gasifier with respect to throughput. A unique feature of the process is that almost 60% of the methane in the product gas is made in the gasifier.

The Methanator

The development of the methanation step has proceeded on two processes: the hot-gas-recycle (HGR) [2] and the tube-wall reactor (TWR). [3] Development

1/ J. H. Field, et al. Pilot Plant Studies of the Hot-Carbonate Process for Removing Carbon Dioxide and Hydrogen Sulfide. BuMines Bull. 597, 44 pp. 1962.

2/ A. J. Forney, R. J. Demski, D. Bienstock, and J. H. Field. Recent Catalyst Developments in the Hot-Gas-Recycle Process. BuMines R.I. 6609, 32 pp., 1965.

3/ W. P. Haynes, J. J. Elliott. A. J. Youngblood, and A. J. Forney. Operation of a Sprayed Raney Nickel Tube Wall Reactor for Production of a High-Btu Gas. Preprint of Division of Fuels Chemistry of ACS Meeting, Sept. 1970, Chi., Ill.

at present is in the TWR system. This system is shown in figure 3. Within the methanator, the gas (fresh feed plus recycle) passes up over the outside of seven 2-inch diameter tubes which are thermal sprayed with Raney nickel to a thickness of 0.020 to 0.030 inch. The tubes are baffled to increase turbulence of the gas flow. Results of the latest test are shown in figure 4. The present test has been operating 2900 hours, equivalent to 240,000 SCF of high-Btu gas/lb of catalyst. The desired sulfur content before methanation must be less than 1 ppm; in methanation tests at Bruceton the sulfur content of the feed gas has been as low as 10 ppb.

NEW DEVELOPMENTS

Pretreatment

The throughput of the gasifier would be improved if a coal coarser than 70% through 200 mesh could be gasified. Attempts were made to pretreat such coals, but nothing coarser than 30% through 200 mesh could be pretreated in the present free-fall unit. The pretreater was changed back to the fluid-bed system used originally. 4, 5/ Figure 5 shows a schematic of the new system. Coal 20 mesh by 0 was pretreated easily with less oxygen than needed for the free-fall system.

Gasification

The change to a coarser coal permitted an increase in the linear velocity; thus the coal feed could be raised equivalent to about 50 lb/hr-ft^3.

A change in the bed height of the gasifier from 6 to 12 feet is scheduled, which would raise significantly the throughput per gasifier internal cross-sectional area.

Methanation

Because the tubes were difficult to change in the reactor shown in figure 3, where the Ranel nickel was sprayed on the exterior of 2-inch-diameter tubes, the reactor was revised. We sprayed the catalyst on the inside surface of 4-inch-diameter tubes. This revised unit is being tested. Since then we have started development of a new method of spraying the catalyst, with the objective of coating the inside of tubes as small as 1-5/8 inch in diameter and as long as 30 feet. Such an improvement now appears feasible because we found we could use a wire spray gun instead of the powder spray gun, which had many limitations.

Pressure

Extrapolation of results of tests at 20, 30, and 40 atmospheres show that pressures of 70 atmospheres should result in a yield of methane exceeding 5 scf/lb coal feed. This higher pressure would reduce the oxygen requirement by one-fourth. The prototype plant will be designed for 70 atmospheres to verify this conclusion.

4/ A. J. Forney, R. F. Kenny, S. J. Gasior, and J. H. Field. Fluid-Bed Gasification of Pretreated Pittsburgh Seam Coal. Preprint of Division of Fuels Chemistry of ACS, September 1966, New York, N.Y.

5/ A. J. Forney, R. F. Kenny, S. J. Gasior, and J. H. Field. The Production of Non-Agglomerating Char From Coking Coal in a Continuous Fluid-Bed Reactor. Preprint of Division of Fuel Chemistry of ACS, September 1964, Chicago, Ill.

CONCLUSION

The Synthane Process steps of gasification, purification, and methanation have been proven at the Bruceton laboratories. Recent improvements of these steps have helped the overall process and should make the process more attractive economically. Although the design of the prototype plant must be fixed soon, continued research is expected to improve the process.

TABLE 1. - <u>Latest test results of the Bruceton gasifier
compared to assumptions made in the
economic report</u>

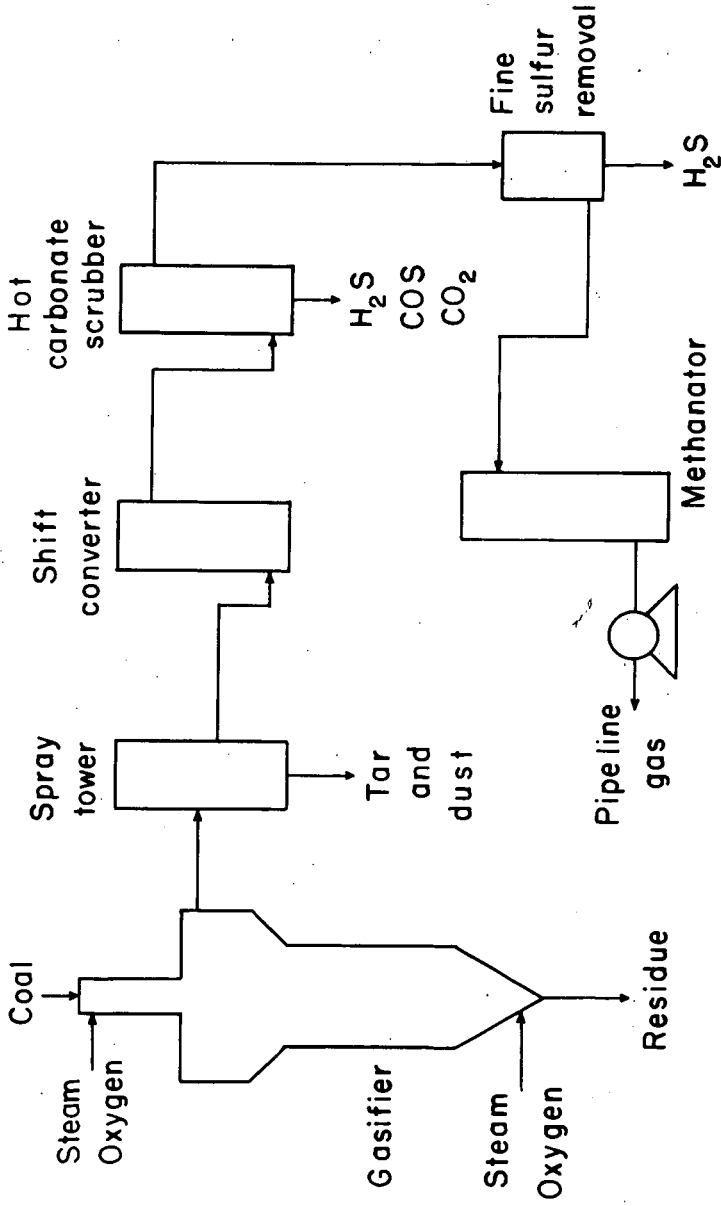| Assumptions	Plant test			
Throughput	Test 196	199	197	204
25 lb/hr-ft^3	32	30	43	36
Product Gas				
16.2 SCF/lb	18.0	18.6	16.2	17.3
18.0 SCF/lb MAF	19.4	20.2	17.5	19.0
CH$_4$				
4.45 SCF/lb	4.40	4.65	4.16	4.51
4.95 SCF/lb MAF	4.74	5.04	4.48	4.97
CH$_4$ 27.5%	24.4	24.5	25.4	25.9
Carbon Conversion				
To Gas 66.8%	66.3	71.0	59.0	64.9
Steam Decomposition 39%	38	32	32	37

Figure 1. System Used to Make High-Btu Gas From Coal
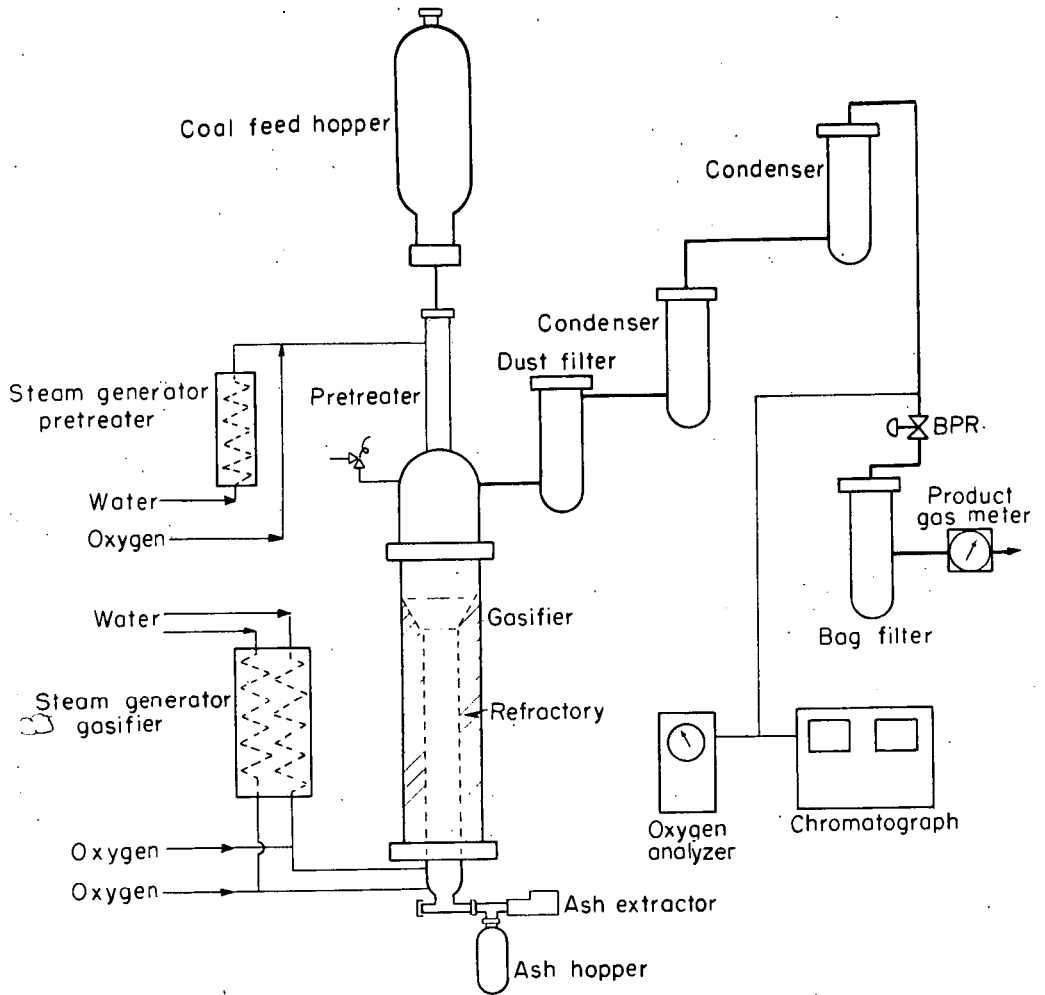
L-11247

Figure 2. 40-Atmosphere Fluid-Bed Gasifier
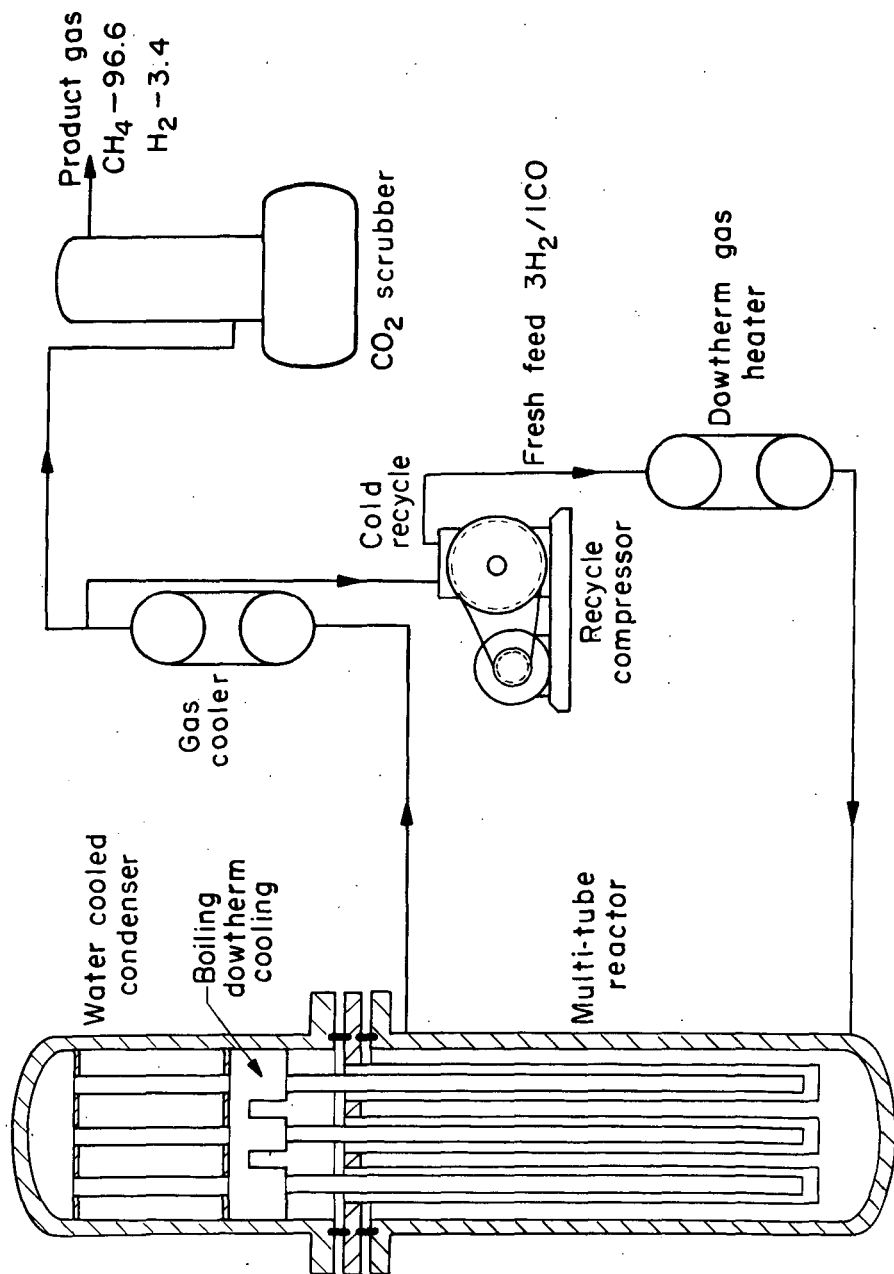(free-fall pretreater)

L-11087

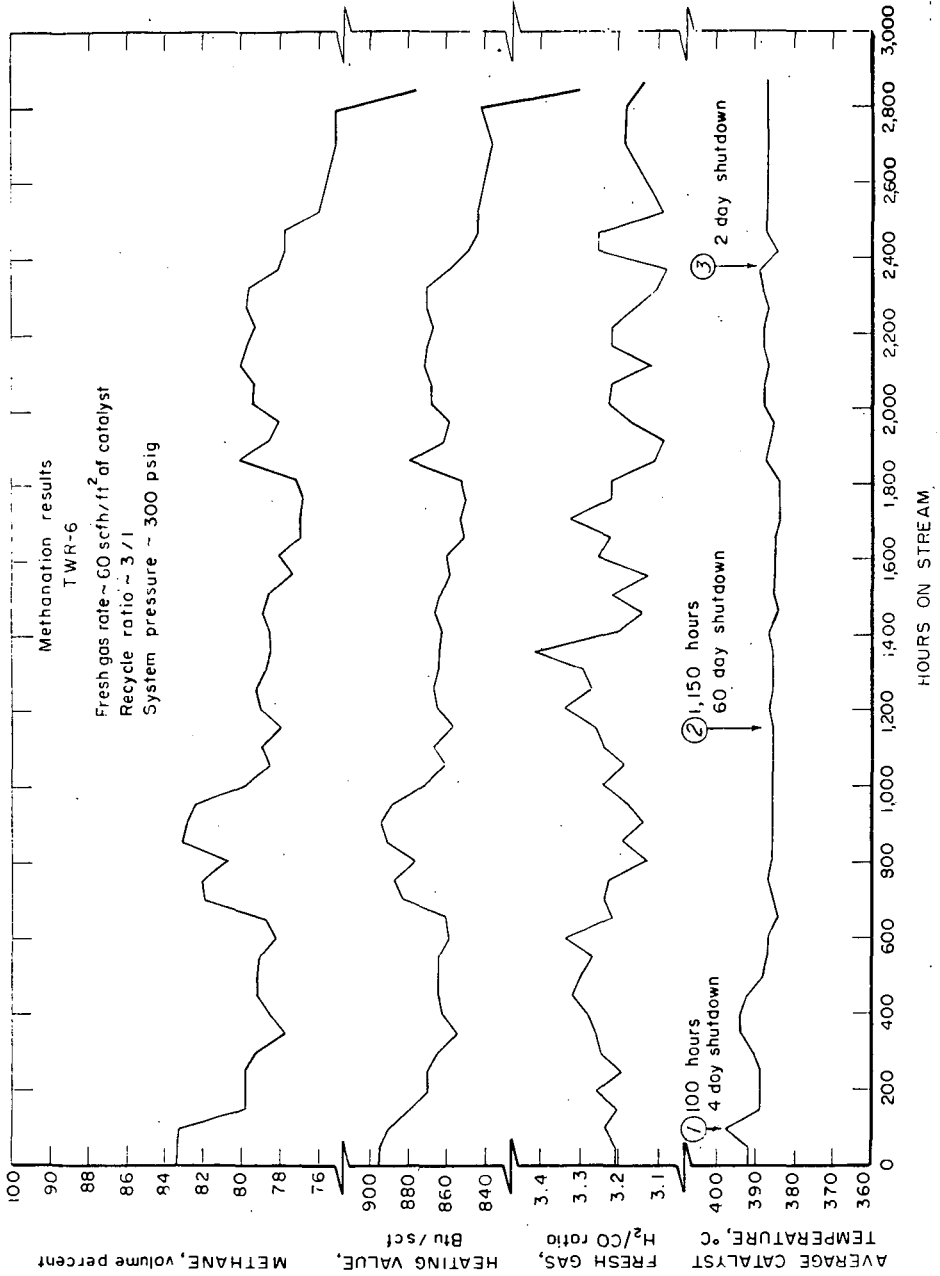Figure 3. Simplified Flowsheet of Pilot Plant for Producing High-Btu Gas
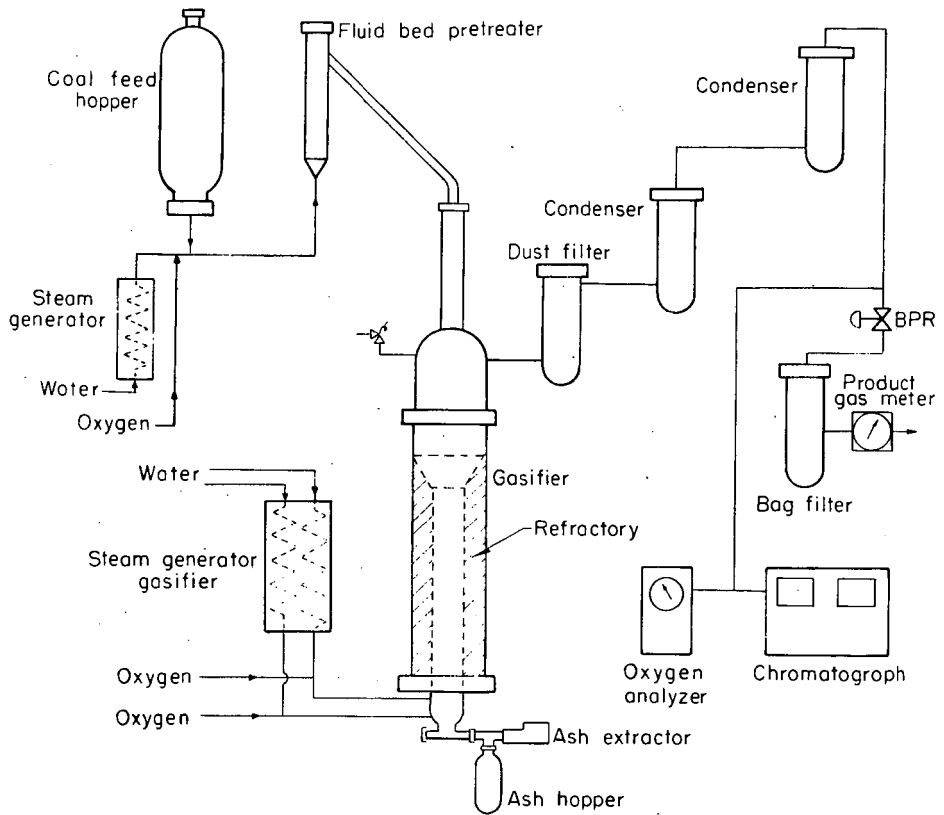
L-11657

Figure 4. Methanation Test Results

L-12 221K

Figure 5. 40-Atmosphere Fluid-Bed Gasifier
(fluid-bed pretreater)

BASIC FEATURES OF THE CO_2 ACCEPTOR GASIFICATION PROCESS

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Consolidation Coal Company, P. O. Box 1687, Rapid City, SD 57701
Consolidation Coal Company, Research Division, Library, PA 15129

INTRODUCTION

The Rapid City plant embodies a unique chemical, fluidized bed system originally conceived by Consolidation Coal in the 1940's. The plant is located where there is easy access to various sources of lignite in the huge Western lignite field. The scale of the plant was chosen to permit obtaining precise engineering data and scale-up information for commercial facilities.

At this point, the laboratory work is complete, plant construction is complete (except for insulation and painting) and preliminary operations have started.

PROCESS DESCRIPTION

The production of pipeline gas by gasification of lignite using the Consol process has four main steps. The overall steps are shown in Figure 1. They are coal preparation, gasification, gas purification, and methanation. Coal pretreatment is also shown; however, this step is unnecessary when lignite is gasified. To handle sub-bituminous coal, it is necessary to use a pretreatment step comparable to that employed in other gasification processes.

The key reactions that take place in the CO_2 Acceptor Process are summarized in Table I. The reactions of carbon, carbon oxides, steam and hydrogen are the same in all gasification processes. The reactions shown that involve calcium oxide, calcium carbonate, and carbon dioxide are unique to the Consol process. Through these reactions, heat is transported between the gasification and combustion sections and simultaneously carbon dioxide is removed from the product gas.

In the coal preparation step, the lignite is crushed to approximately 8 mesh in a mill and simultaneously dried from a total moisture content of 35% to 5% using hot gas.

As a final step in preparation, lignite is preheated in a fluidized bed to 572°F, which also drives off some bound moisture and possibly some CO_2. This step will be modified to a pretreatment when sub-bituminous coal is used.

Lignite is a non-caking coal; and as a result, the dried and ground material leaving the fluidized bed heater is ready for gasification.

The gasification section of the process, as incorporated in the pilot plant, is shown in Figure 2. The crushed and dried lignite powder is introduced to a lockhopper system. Pressure in the hopper is increased to gasification pressure of 150 to 300 psig, and the powder is then fed through a rotary feeder into the first reaction vessel called the devolatilizer.

In the devolatilizer, the lignite reacts with a hydrogen-rich stream from the gasifier at 1500°F. The introduction point of lignite is at the bottom of the bed and the volatile matter from the lignite leaves the top of the bed as methane, carbon monoxide, and hydrogen. The configuration of the vessel and operation conditions are such that there is essentially no condensable hydrocarbon in the product gas.

The devolatilized lignite (which we can now refer to as char) flows by gravity from the devolatilizer. It is then lifted into the gasifier (the vessel on the right in Figure 2) using superheated steam as a motive gas. In the pilot plant, we have the flexibility of recycling gas to the gasifier through a cleanup and compressor system. This is also true of gases from the other reactors. This capability is included in the pilot plant to permit studying alternative operating schemes.

Carbon in the char is gasified by reaction with steam in the gasifier. Residual char, containing about 35% of the original carbon in the lignite, flows out of the gasifier by gravity and is lifted pneumatically to the regenerator using a very small stream of gas recycled from the regenerator as the motive gas.

In the regenerator, the residual carbon is burned at approximately 1900°F to supply the heat needed for the reversal of the carbonation reactions that take place in the devolatilizer and gasifier vessels (see Table I). The residual ash, containing about 5 absolute per cent of the original lignite carbon content, is entrained in the outlet gas from the gasifier. This residue, together with fines from the dolomite cycle which will be described next, is removed from the regenerator offgas in a cyclone. In the pilot plant at Rapid City, these solids will be lockhoppered out of the system into a tank where they will be slurried with water and pumped to a settling pond.

The dolomite cycle carries heat from the combustion in the regenerator to the gasification and devolatilization as was mentioned earlier. This is the key to the CO_2 Acceptor Process. Thermodynamic considerations associated with this cycle will be described later. In the regenerator vessel, calcium carbonate is calcined to calcium oxide adsorbing heat, and this heat is released when the calcium oxide is converted back to calcium carbonate by reaction with CO_2 in the other reactors.

The regenerator runs essentially full of fluidized dolomite. Char from the gasifier enters the bottom and is elutriated out the top after combustion with air. Calcined dolomite from the regenerator flows into the other two vessels by gravity.

In the gasifier and the devolatilizer, the top bed is essentially all fluidized char. Dolomite is introduced at the top and comes out of the char bed from a second fluidized bed in the small section at the bottom of each vessel. The small sections are called boots. There is little char in the dolomite boot section.

Dolomite from these boots flows by gravity to an engager pot from where it is lifted back into the regenerator. Air will be used as the motive fluid in commercial plants. The pilot plant is also equipped to use recycled gas from the outlet of the regenerator.

The product gas finishing and methanation steps are not included in the Rapid City Pilot Plant. The techniques will be similar to those used for other gasification processes.

STATUS

At this point, bench-scale research on the CO_2 Acceptor Process is finished and the pilot plant is complete except for insulation and painting. Some components of the plant, mostly standard designs, have been tested. Training of operating personnel will be completed by mid-October, at which point dry runs will begin. These runs will test the gasification controls, the special mechanical features, and the fluo-solid transfer between the reactor beds. These tests represent a start at completing the future developments numbered "1" and "2" in Table II.

We expect to begin gasification starting in February, and at that time, we will begin acquiring knowhow for commercial operation (Item 3 in Table II).

Presently the plans for the pilot plant involve testing the least severe process concepts first and gradually working toward more difficult combinations. Each test will bring us closer to achieving the three future developments in Table II.

GENERAL REMARKS REGARDING OPERABILITY PROBLEMS IN GASIFICATION PROCESSES

*   The price for synthetic gas from coal is highly influenced by charges related to the investment of the plant. This capital intensity is dependent on the nature of the feedstock.

As compared to petroleum or the usual petrochemical raw materials, coal has properties which uniquely influence a processing plant investment. Some of these properties are:
1. Coal is a solid with varying and only partially predictable thermoplastic properties.
2. It contains mineral matter of varying composition.
3. It has varying chemical and petrographic compositions, and an undefined complex chemical structure.
4. It is thermally unstable and decomposes to yield a wide spectrum of gases, oils, and solids, the properties of which are influenced by the conditions of pyrolysis.
In addition to the problems created by these unique raw material properties, there is the basic necessity of supplying the large quantities of heat required for the endothermic gasification reaction at the high temperature levels needed for acceptable reaction kinetics.

Thus, the following potential operational problems are common to all coal gasification processes:
1. Feeding.
2. Caking.
3. Handling of volatile matter.
4. Supply of process heat.
5. Deposit formation.
6. Ash removal.

In the evaluation of chemical processes, the judgment usually culminates in a comparison of the projected process economics. When this is done for commercially proven processes, the operability is already established and the result of such an evaluation is reliable.

For conceptual processes, however, with only bench-scale results available, judgment of alternative processes using cost estimates is a rather risky venture, especially in the field of coal gasification.

While the individual concept may feature a different degree of difficulty in each of the problem points listed above, it is obvious that processes with the most straight-forward solution to these problems have the best chance to become commercialized. In judging concepts, one should, therefore, examine the chances for operability first of all. Many concepts based on sound chemistry have failed because some of the problems have turned out to be insurmountable.

The CO_2 Acceptor Process will be examined in the following paragraphs in each of the areas that create operational problems.

FEEDING PROBLEMS

A problem exists whenever a large quantity of solids has to be introduced at a continuous and controlled rate into a pressurized high temperature reactor. The three principal means for accomplishing this step are a lockhopper system, pumping coal as a slurry, or a piston feeding device.

The lockhopper system was chosen for the CO_2 Acceptor Process for the following reasons:
1. Operability commercially established.
2. Low investment and operating cost for the pressure level desired.
3. Carbon dioxide can be used for pressurizing.
4. Amenable to automation.

In the pilot plant, the system consists of two hoppers, each with a capacity of one hour operation at 1.5 tons (Figure 3). The hoppers are operated alternately. While one of them is connected with the system and feeding, the other one is blocked off while it is depressurized, filled with coal, and finally repressurized with CO_2.

Operation is semi-automatic by electrically operated valves and push buttons. The feedrate is controlled from the main control room by variable speed rotary feeders, and the amount is measured by a system of loadcells and transmitters.

The lockhoppers are continuously purged with a small flow of CO_2 to keep hot process gas out during the feeding period and to prevent air from entering during the filling operation. Total CO_2 required for pressurizing and purging, 1300 SCFH, is small compared to other processes. The reason for this is that the lockhoppers are operated with 500 to 600°F preheated coal which tends to decrease pressurizing and purge gas requirements.

CAKING PROBLEMS

The CO_2 Acceptor Process is primarily designed to process lignite and sub-bituminous coals. While the lignite does not present a problem, normally non-caking sub-bituminous coal displays weak caking properties at high partial pressures of hydrogen. Although we do not expect agglomeration of the fluid bed, the "popcorn"-shaped (cenospheres) char produced by pyrolysis of caking coals can cause operational problems.

The plant is equipped to pretreat caking coal by operating the fluidized preheater at oxidizing conditions.

The preheater system (Figure 4) of the pilot plant is designed for atmospheric pressure. It is sized for eight hours retention time of the coal in order to serve as a surge between the drying system and the gasification section.

The system consists mainly of a natural gas-fired furnace, a fluidized bed vessel, and a venturi scrubber. In operation as an oxidizer, the furnace is fired with excess air. As tempering gas, we have the option to use either steam or flue gas from the regenerator of the gasification section. The outlet gas is quenched and scrubbed in the venturi scrubber and released to the atmosphere.

Because of the eight hour retention time, mild oxidizing conditions are sufficient. It is intended to operate at 500 to 600°F and up to 6% O_2 in the inlet gas with sub-bituminous coals.

In a commercial plant, this system would be modified for operation at a shorter retention time, and consequently, stronger oxidizing conditions.

HANDLING OF VOLATILE MATTER

Each coal evolves volatile matter during thermal treatment. The amount and the composition of these volatiles are determined by the rank of the coal as well as by the conditions of this treatment. In addition to fixed gases, hydrocarbons ranging from heavy tar to light distillates of aliphatic or aromatic nature are usually present as vapors in the raw gas.

Separation of these condensables from the raw gas which usually carries more or less dust can present a problem. The degree of difficulty and the numbers of steps required for cleanup is in proportion to the range of the hydrocarbon spectrum present in the gas.

We came to the conclusion that liquid hydrocarbon byproducts should be avoided by all means if the process configuration provides the possibility. Even though they have a high BTU price, the return would neither justify an additional process complication nor the investment for additional separation and refining equipment.

It was found that a minimum temperature of 1500°F and a retention time of the vapors in the reactor bed of at least 17 seconds in the presence of steam and hydrogen would break the volatile matter down to permanent gases, mainly methane, a trace of ethane, and some carbon oxides.

The devolatilizer reactor was designed for these conditions. Fresh coal is fed into the bottom of the fluidized bed, and the bed height provides sufficient retention time for the ascending vapors to react with steam and hydrogen present in fluidizing gas.

As a result, our gas clean-up system is rather simple (Figure 5). It consists of a quench tower with disc and doughnut internals, a quench separator, and a venturi scrubber.

The quench water is recycled to the tower from the separator and indirectly cooled. Excess water from condensation of undecomposed steam carries the precipitated solids out of the system. We have provided a baffle in the quench separator to handle light oil which might occur at upset conditions or startup. In the venturi scrubber, traces of dust are removed from the gas before it enters the recycle compressor.

As mentioned before, our gas treatment is limited to these steps because we did not install equipment for methanation and associated preparatory treatment.

SUPPLY OF PROCESS HEAT

The method of heat supply has a great impact on the economy of a gasification process. Problems are usually associated with the requirement to supply the heat at relatively high temperature levels to the reaction zone.

If consideration is limited to systems producing essentially nitrogen-free water gas, coal gasification processes can be categorized in terms of the methods of supplying heat to the gasification zone; i.e.,

1. Oxygen-blown systems.
2. Cyclic processes.
3. Indirectly heated processes (electrical heat, nuclear energy, etc.).
4. Heat carrier processes.

The CO_2 Acceptor Process falls in the last category. However, it is distinguished from other solids recirculation processes by the fact that most of the heat is liberated by chemical reaction.

As described before, calcined dolomite at 1950°F is introduced to the top of the reactor beds in granular form. Because of their higher gravity, the particles shower through the beds. On their way down, they react with CO_2 which is always present in devolatilization and gasification zones.

At the bottom of each reactor, the carbonated dolomite is collected in a bed of smaller diameter and a higher fluidizing velocity. From these small beds it is transferred to the regenerator for calcination at 1950°F (see Figure 2). This reaction requires heat which is supplied by combustion of residual char from the gasifier. After this treatment, the dolomite is ready for another cycle.

The average acceptor particle will undergo about 50 cycles of calcining and recarbonation until it is replaced by dolomite makeup. This replacement rate of 2% is necessary to maintain a reasonable activity level in the circulating inventory. We expect that under these conditions between 30 and 40 mol per cent of the circulating dolomite will participate in the recarbonation reaction.

Figure 6, a T-H diagram, shows that only 27.5% of the heat carried with the dolomite is sensible heat and that the major portion of 72.5% is heat of reaction. This diagram is for one mol of dolomite at 40% activity and 1950/1500°F temperature differential. The total heat supplied to the process by one mol of dolomite under these conditions is about 40,000 BTU.

About three and one-half times as much dolomite would be required at the same temperature differential for the same heat supply if all the heat were supplied by sensible heat. All solids which have been used in this service as heat carriers such as sand or char, etc., have roughly the same specific heat as dolomite. In the CO_2 Acceptor Process, the weight ratio of circulated dolomite to coal fed is between 3 and 4; whereas other gasification processes with chemically inert heat carriers have to circulate a burden of 10 to 15 times their coal feed. Since increased solids flow in a process means more investment, the acceptor process should be more economical than other processes of the heat carrier category.

DEPOSIT FORMATION

Deposit formation is a problem common to all gasification processes. Considerable time was spent in studying this problem in the CO_2 Acceptor bench-scale studies. It was found that deposits can occur in all three reactors. The mechanism of formation is different in each process vessel.

Devolatilizer deposits are formed by interaction between sodium sulfate, pyrite and SiO_2, while in the gasifier only the last two components are involved. There are no obvious means of preventing these deposits. The bench-scale data indicates, however, that with most Western coals, commercial operating periods of at least six months can be achieved without descaling the reactors. Obviously, however, coals having high sodium sulfate and pyrites contents should be avoided. In passing, it should be noted that the devolatilizer and gasifier deposits are not related to the presence of the dolomite acceptor; all gasification processes utilizing Western coals will encounter this problem.

In contrast, the deposits found in the regenerator are unique to the CO_2 Acceptor Process and can be much more severe. It was found that the formation mechanism is based on sulfur reactions in regeneration and that the deposition can be prevented by controlling the conditions in the regenerator.

Calcined dolomite is as good an acceptor for sulfur compounds as for CO_2. The sulfur forms CaS, mainly in the devolatilizer but also in the gasifier. In the regenerator the CaS content of the acceptor is oxidized in the $CaSO_4$ by either of the following reactions:

$$1/4 \; CaS + CO_2 \longrightarrow 1/4 \; CaSO_4 + CO$$
$$or \quad 1/4 \; CaS + 1/2 \; O_2 \longrightarrow 1/4 \; CaSO_4$$
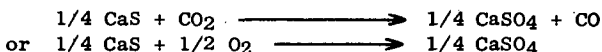
When CaS and $CaSO_4$ exist together in the absence of carbon oxides, sulfur is rejected and the calcium is converted to CaO by the reaction:

$$3/4 \; CaSO_4 + 1/4 \; CaS \longrightarrow CaO + SO_2$$
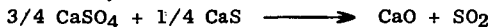
This last reaction is the sum of various intermediate steps. As an intermediate in the reaction, at temperatures above 1750°F, a transient liquid phase occurs which collects and solidifies on the regenerator walls.

In studying the problem, it was found that a slight partial pressure of carbon monoxide in the regenerator would prevent the diffusion of the transient liquid to the surface of the acceptor particles and thereby avoid its deposition on the walls of the vessel. The amount of CO required depends primarily on the total amount of sulfur fed to the regenerator and must be determined individually for each coal or set of operating conditions.

There is a price tag associated with this solution; namely, the rejection of carbon in the regenerator, which means loss in overall efficiency. Approximately 5% of the original carbon of the coal must be sacrificed.

ASH REMOVAL

This problem generally involves three steps:
1. Separation of ash from the process.
2. Removal from the pressure system.
3. Disposal.

Separation of ash from the process takes place in the regenerator. The particles, while reacting with air, move upwards through the interstices of the dolomite bed. The ash particles are stripped out of the bed because they are much finer than dolomite particles.

Separation of entrained ash from the gas takes place in a cyclone and the rest is removed in a wet scrubber. Before entering the cyclone, the gas/solid mixture is cooled from 1900 to 1000°F by means of a water-jacketed pipe.

The removal from the pressure system is accomplished by alternately operating lock-hoppers of a configuration similar to those described for coal feeding.

The residue from the lockhoppers contains about 80% of the sulfur content of the feed coal. Because it is present as CaS, this material cannot be disposed of by dumping Rain would gradually liberate H_2S causing a bad odor in the vicinity of the dump area.

In the pilot plant, the ash lockhoppers are discharged into an agitated tank and slurried with water. The slurry is reacted with CO_2 to release H_2S which is incinerated The innocuous solid residue is then pumped into a settling pond.

Disposal in a commercial plant would be accomplished in a similar manner, except that the liberated H_2S would be converted to sulfur. Investigations are under way to find commercial use for the residue.

CONCLUSION AND SUMMARY

The construction of the Rapid City Pilot Plant is almost completed. After initial cold runs, gasification tests will be started in February of next year.

The process was examined with respect to the six main problems commonly associated with coal gasification.

For the problems of feeding, caking, and ash removal, the process configuration offers the possibility of straightforward solutions, while the problem with condensable hydrocarbons can be eliminated completely by appropriate process steps.

The process embodies a unique solution to the heat supply problem wherein the heat is transported chemically. The required solids circulation rate is only a fraction of that which would be required if all the heat were supplied as sensible heat.

The formation of ash deposits in the devolatilizer and gasifier is expected with Western coals in all gasification processes. Bench-scale data indicate that with most Western coals, commercial operating periods of at least six months can be achieved without scale removal.

Deposit formation in the acceptor regenerator is a potentially serious problem. The problem was eliminated in bench-scale tests at the expense of a loss in process efficiency.

TABLE I

PERTINENT REACTIONS IN CO_2 ACCEPTOR PROCESS

	HEAT OF REACTION $\underline{BTU/MOL}$ at 298.16° K
$C + H_2O\ (g) \longrightarrow CO + H_2$	56,500 Endothermic
$CO + H_2O\ (g) \longrightarrow CO_2 + H_2$	17,700 Exothermic
$CaO + CO_2 \longrightarrow CaCO_3$	76,200 Exothermic
$CaCO_3 \longrightarrow CaO + CO_2$	76,200 Endothermic
$2C + H_2 + H_2O\ (g) \longrightarrow CH_4 + CO$	24,300 Endothermic

TABLE II

MOST SIGNIFICANT PAST AND FUTURE DEVELOPMENTS

<u>PAST</u>

1. Concept of Using Acceptor in Commercially Feasible Process

2. Correlations for Dolomite-Char Fluid Systems and for Reaction Rates

3. Methods for Avoiding Agglomeration and Slagging

<u>FUTURE</u>

1. Confirmation of Practical Operability and Plant Capacity

2. Obtain Information for Precise Engineering Scale-Up

3. Acquire Knowhow for Commercial Start Up and Operation

PIPELINE GAS FROM COAL

```
┌──────────┐                                    ┌──────────┐
│   COAL   │                                    │ PIPELINE │
│          │                                    │   GAS    │
└──────────┘                                    └──────────┘


         SOLIDS HANDLING                              GAS FINISHING
   ┌──────┬──────────┬──────┐  ┌──────────┐  ┌────────┬────────┬──────────┐
   │ COAL │   COAL   │ COAL │  │          │  │        │        │ DEHYD'N &│
   │ PREP.│ PREHEAT  │PRESS.│  │GASIFICATION│ │PURIF'N │METH'N  │COMPRESS'N│
   └──────┴──────────┴──────┘  └──────────┘  └────────┴────────┴──────────┘
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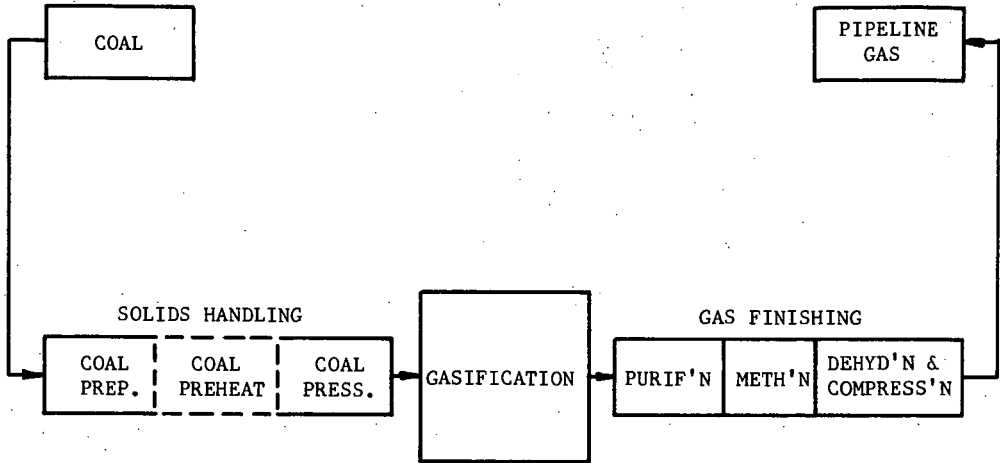
FIG. 1

CO$_2$ ACCEPTOR PROCESS

RAPID CITY PILOT PLANT- GASIFICATION SECTION

ASH & ACCEPTOR FINES

GAS COOLING & CLEAN-UP

TO RECYCLE & VENT

GAS COOLING & CLEAN-UP

REGENERATOR 1870-1940°F

GAS COOLING & CLEAN-UP

TO SULFUR REMOVAL & METHANATION

DEVOLATILIZER 1500-1525°F

GASIFIER 1500-1625°F

AIR

LIGNITE

INERT GAS

INERT GAS

STEAM & RECYCLE GAS

STEAM & RECYCLE GAS

FIG. 2

LOCK HOPPER SYSTEM

LIGNITE FEED

VENT

LOCK VALVES

VENT

PURGE CO_2

LOCK
HOPPER

LOCK
HOPPER

PURGE CO_2

FEEDER

LOCK
VALVES

FEEDER

LIGNITE TO REACTORS

FIG. 3

PREHEATER SYSTEM

NATURAL GAS

COMBUSTION AIR

VENT

LIGNITE
PREHEATER
FLUID BED

WATER

VENTURI
SCRUBBER

FURNACE

FINES
SLURRY

FLUE GAS

STEAM

FIG. 4

GAS CLEAN-UP SYSTEM AT PILOT PLANT

NET MAKE GAS TO
THERMAL OXIDIZER

CLEAN GAS
TO RECYCLE
COMPRESSOR

VENTURI
SCRUBBER

QUENCH
TOWER

SUCTION
SCRUBBER

GAS FROM
REACTOR

COOLER

SEPARATOR

TO SLOP
OIL TANK

TO SETTLING
POND

PUMP
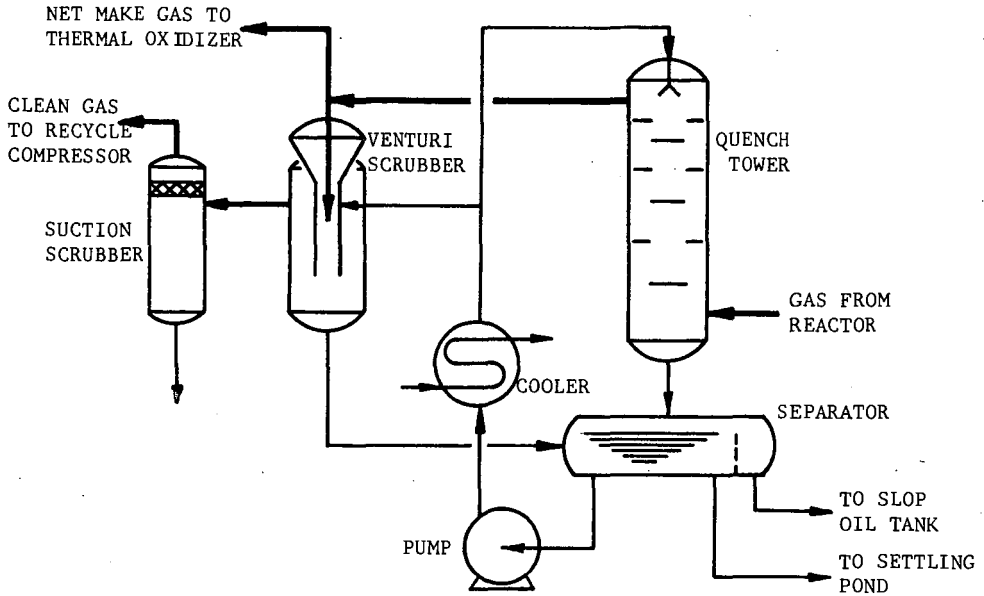
FIG. 5

HEAT SUPPLY OF ACCEPTOR AT 40% ACTIVITY

ENTHALPY ΔH (M BTU)

TEMP. T
(°F)

1950
1900

1800

1700

1600

1500

MgO·CaO

MgO·CaO + CO_2

40% MgO·$CaCO_3$
60% MgO·CaO

10   20   30   40

11,000   29,000 BTU

27.5%   72.5%
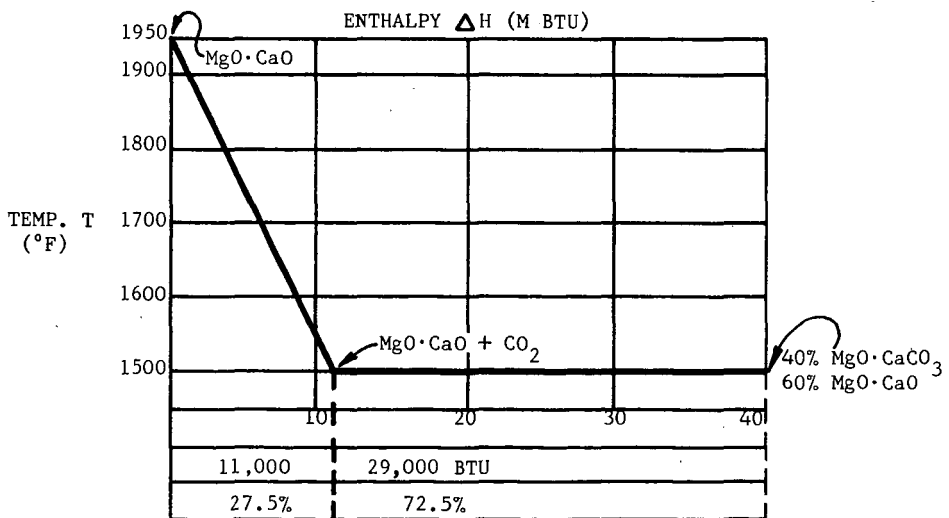
FIG. 6

STRONGLY CAKING COAL GASIFIED IN A STIRRED-BED PRODUCER

By

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INTRODUCTION

Partial combustion of coal with continuous addition of steam converts a large fraction of the energy in the solid fuel into combustible gases: carbon monoxide, hydrogen and hydrocarbons plus a small fraction of non-combustible carbon dioxide. This gasification process is well known, and it has been practiced on a large scale for many years. It is carried out in a retort-type vessel called a producer, and the product is known as producer gas.

Gasification with air dilutes the gas with inert nitrogen to the extent of about 50 percent. This dilution can be reduced or eliminated entirely by substituting oxygen for a part or all of the air. Process requirements and economics for each application determine whether air or oxygen will be used.(1)[1]

Gas producers are usually classified in three categories according to the characteristics of the fuel bed, namely: fixed, fluidized, or entrained. Within this broad description, many innovations have been proposed. Sixty-five producers of all types are described in a recent report.(2) The fixed-bed producer is the only type accepted thus far for commercial use. Fixed-bed producers reached their maximum use in the United States about 1925 when approximately 11,000 were in service in the United States. They are still used to a minor extent in Europe, South Africa, Australia, and Japan for firing industrial furnaces and generating town and synthesis gas.

Interest in pressurized gas producers is increasing here and abroad for application to thermal power generation using combined gas and steam turbines with gas cleaning proceeding combustion. The first commercial plant of this type is scheduled for completion the summer of 1971. Its output is 170 Mw, and the location is Lünen, West Germany. The pressurized gas producer also has potential for the production of high-Btu pipeline gas from coal because of the substantial amount of methane formed during gasification.

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[1]/ Numbers in parenthesis refer to references at end of paper.

The U.S. Bureau of Mines is undertaking the development of a fixed-bed producer at its Morgantown, W.Va. Energy Research Center.(3) This experimental producer provides more extensive stirring of the fuel bed than has been the practice heretofore. It conforms to the accepted concept of a fixed-bed producer in all other respects.(4) The designation stirred-bed producer is proposed in order to distinguish this type from the others.

The fixed-bed producer has demonstrated thermal efficiencies (percentage of heating value of the fuel appearing in the gas) of about 90 percent for hot raw gas and about 75 percent for cold clean gas. Generally the fuel has been limited to coke, anthracite, or noncaking bituminous coals. Mildly caking coals have been used in some commercial producers which were provided with a mechanical stirrer for agitating the top level of the bed. This stirring action broke the weak coke that formed and filled voids in the bed. The bulk of the bituminous coal located in the eastern half of the United States is too strongly caking to be gasified in a fixed bed. A strongly caking coal from West Virginia's Pittsburgh seam was gasified in tests performed at Dorsten, Germany, in a fixed-bed producer equipped with a top stirrer. The caking property had to be reduced by adding ash from local coal in order to prevent adherent coke from stopping the coal flow.(5) A coal's performance under conditions existing in a fixed bed varies with its plastic properties, mineral content, heating rate, particle size, and operating conditions. Development of equipment and techniques for gasifying caking coals in a fixed bed will provide a versatile, economical system for converting coal to gas.

EXPERIMENTAL

Equipment

The stirred-bed producer is shown in figures 1 and 2. It operates at pressures to a nominal 100 psia. Inside dimensions are 3.5 feet in diameter and 24 feet in length. The fuel-bed agitator or stirrer is the design feature that distinguished this experimental producer from other producers of fixed-bed design. The agitator consists of a vertically mounted rotating shaft to which is attached two horizontal arms, or rabbles, located 2-feet apart. Both rabble arms and the entire length of the shaft are cooled by circulating water. A third rabble, uncooled and positioned 2-feet above the middle arm, levels the top of the bed. The variable speed drive acts through two lever arms and ratchet mechanisms which rotate the shaft and move it vertically in reciprocal motion. The distance traveled vertically is controlled by the positioning of limit switches within a maximum span of 6-feet 4-inches. For the experiments reported here, the vertical travel was limited to 2-feet. Settings of 2 or 3 on the variable speed drive gave agitator rates of 15 or 22-1/2 revolutions per hour and 3-1/2 or 5-1/4 feet per hour travel, respectively. Limits in rotation were 8 to 40 revolutions per hour and in vertical travel 2 to 9-1/3 feet per hour. A setting of 3 was used most of the time during these tests.

Continuous infrared analyzers record the concentration of carbon monoxide and carbon dioxide in the product gas. Continuous monitoring of

the fuel bed for bed depth, occurrence of voids, location of combustion zone, and presence of excess ash is obtained by nuclear instruments, figure. 3. The percentage of gamma-ray radiation transmitted through the vessel from a cobalt-60 source varies with the thickness and bulk density of the material through which the gamma-rays pass. Coal, ash, and gas attenuate the radiation in varying degrees dependent upon their presence or absence and bulk density, but attenuation by the vessel wall is constant.(6) A 625-millicurie source with three detectors locates the top of the fuel bed. Coal is added when the middle detector indicates "empty". A 4-curie source and two detectors with outputs recorded on a strip chart shows the presence or absence of voids or high-ash level as indicated in figure 4. Adjustments to the stirring rate are based on this data.

Operation

The fuel bed moves downward countercurrent to the ascending hot gases resulting from partial combustion of the coal. Air and steam are admitted below the grate. Fuel consumption is controlled by adjustment of the air rate, and temperature is controlled by adjustment of the steam rate. The temperature is held below the ash fusion temperature to avoid forming clinkers which clog the grate.

Coal is fed intermittently to the top of the bed by adding batches weighing 200 - 250 pounds about every 10 minutes through lock hoppers pressurized with inert gas. Ash is discharged by means of a rotating grate and removed through a lock hopper. The removal rate is adjusted by varying the grate rotation between 4 and 10 rph. The combustion zone is held about one foot above the grate. The normal bed depth above the grate is about eight feet. The interface between the ash and combustion zones is generally below the zone detected by the nuclear density gage; detection is by means of thermocouples having vertical spacing of 12 inches. Temperature differentials give a good indication of the interface location, although the measured temperatures are much lower than those in the interior of the bed because of the close proximity of the thermocouples to the water-cooled wall. A 5-foot length of the wall above the top of the grate is water jacketed with the remaining wall area faced with insulating brick.

Gases leave through a side outlet in the top cover, and are reduced in pressure by a control valve. The gas is cleaned of the large part of its dust load by flowing through a one-stage cyclone separator before it is burned in the atmosphere. The gas is continuously analyzed for carbon monoxide and carbon dioxide but is not sampled for steam, tar, or dust. Periodic samples are taken for laboratory analyses of the gas, ash, and cyclone dust.

Start-ups are made with anthracite in order to avoid depositing large amounts of tar in the cold producer. Bituminous coal feed starts when the exit gas temperature reaches 1,000° F. At steady conditions, the gas temperature reaches a maximum of about 1,200° F. About 1.5 tons of coal fill the producer, and one type of coal completely displaces the other in about four hours.

Operations are scheduled on a 3-shift, 5-day week basis. Light off and burnout each require one shift. Holding in banked condition over a weekend is possible, but burnout is the usual practice. These procedures give a 4-day operating period.

Data are presented for strongly caking high-volatile A bituminous, Pittsburgh bed, Monongalia County, West Virginia, and for moderately strong caking high-volatile B bituminous, No. 6 seam, Jefferson County, Illinois. Proximate and ultimate analyses are given in table 1. The Pittsburgh bed coal was cleaned and of 2 x 1-1/4 inch screen size. The Illinois No. 6 coal was cleaned and of 1-1/2 x 3/4 inch size.

Table 1. - Analyses of high-volatile coal
(as received)

	W. Va. Pittsburgh seam[2]	Illinois No. 6[3]
Ultimate		
Carbon	76.9	70.9
Hydrogen	5.5	6.4
Nitrogen	1.3	1.7
Sulfur	2.4	1.2
Oxygen[1]	6.2	14.4
Ash	7.7	5.4
Proximate		
Moisture	1.3	6.9
Volatile matter	36.8	36.1
Fixed carbon	54.2	51.5
Ash	7.7	5.5

1/ By difference
2/ FSI 8, HV 13,850 Btu/lb.
3/ FSI 4-1/2, HV 12,050 Btu/lb.

RESULTS

Process conditions and performance data are given in table 2. These data are the average results from 2 to 4 nonconsecutive operating periods of 7 to 10 hours duration. The results do not represent steady state data, and are shown because they approximate what would be possible. Steam, tar, and dust contents of the gas are not known but can be estimated.

Temperature of the air-steam mixture at the inlet range from $230°$ to $260°F$ and producer gas at the outlet ranges from $900°$ to $1,200°F$. The weight of air is approximately 6 times the steam weight. Air is supplied at about $200°F$ while steam is reheated to about $400°F$, $(75°$ superheat), thus the heat input is approximately 5 percent greater than it would be if combustion of coal were the only source.

A cooling water temperature increases about $25^{O}F$ for the stirrer and $28^{O}F$ for the shell jacket. Water flows for the stirrer and jacket are approximately 310 and 45,600 pounds per hour, respectively. The heat loss to cooling water for the stirrer is virtually nil and for the jacket ranged from 6 to 10 percent of the heat input.

Table 2. - Process conditions and performance averages

	W. Va. Pittsburgh seam		Illinois No. 6 seam
	Test 1	Test 2	Test 3
Data period, hrs. (cumulative)	16	31	36
Pressure, psig	80	80	80
Feeds,			
coal, as rec.[1]/, lb/hr	1,150	1,390	1,630
coal, lbs/Mscf gas	15.7	15.4	18.1
steam, lb/hr	560	720	630
air, lb/hr	3,830	4,600	4,600
Feed ratios, lb/lb			
steam:coal[1]/	0.49	0.52	0.38
air:coal[1]/	3.32	3.31	2.82
Gas yield			
M scf/hr	73	90	90
M scf/hr-sq ft[2]/	7.6	9.4	9.4
scf/lb coal	63.5	64.8	55.2
Gas composition, mol pct[3]/			
CO	20.0	20.5	21.7
H_2	15.5	15.6	15.8
CO_2	7.2	8.7	6.8
CH_4+	2.8	2.4	3.0
N_2	54.5	53.2	52.7
Heating value, Btu/scf (gross)	142	145	164
Heat Losses (estimated), pct of input	13	10	11
Cold gas efficiency, pct	62	65	72

1/ as received
2/ Grate area, 9.6 sq ft.
3/ Dry basis.

DISCUSSION

Performance

Experience with Pittsburgh seam coal indicates that gasification of strongly caking coals can be accomplished by stirring the bed to break coherent coke formations. The compound rotation and axial movement of the agitator provides slow continuous stirring that fills voids and keeps the

bed permeable. The condition of the bed is determined by observing the gas quality and bed density as recorded by control instruments. Gross changes in bed density are immediately apparent and gas quality deteriorates or improves with a respective decrease or increase in density. The bed density gage has great value because it provides a definite and immediate indication of conditions in the bed. The gas analyzers have a time lag, and coal additions produce minor variations in gas composition. This density gage is mounted in a fixed position; it would have greater usefullness if it were mounted on a movable platform traversing the entire bed and the movement controlled from a remote station. A correlation between the stirring rate and gas quality was not obtained, but one probably exists as a slow stirring rate allows voids to be retained for a longer time before they collapse.

A predicted life for the agitator and grate cannot be made at this time. Based on last year's experience, long life can be anticipated. Chances of overheating either agitator or grate are virtually eliminated by controlling operations through the use of instruments. It does not appear to be necessary to extend the stirring into the oxidation zone, and the lower rabble arm is not subjected to high temperature in an oxidizing atmosphere.

Gas Quality

Gas quality is fairly typical for an air-blown producer except that carbon dioxide is 2 to 3 percent higher and carbon monoxide plus hydrogen is lower by the same amount than is normal for a commercial producer. This is attributed to gas temperatures being somewhat lower because the smaller experimental producer has more water-cooled surface per volume of reaction space. Lower gas temperatures in the reduction zone slows the rate of the reaction $C + CO_2 = 2 CO$ and some CO_2 passes through the zone unreduced, expecially if the bed depth is shallow. Design geometry has another effect. The greater wall surface to reaction volume provides better opportunity for unreduced carbon dioxide and steam to leak around the fuel bed.

Heating Value

Gross heating value of the dry gas is slightly higher for Illinois No. 6 seam than for W.Va. Pittsburgh seam. The gas from Illinois coal contained 0.5 percent $C_3H_8 + C_3H_6$ to 0.1 percent for the latter. Heating values are in the range 140 to 165 Btu per scf, which is normal for air-blown producer gas.

Efficiency

The cold gas efficiency is the potential heat in the gas expressed as a percent of the heat input. Two tests with Pittsburgh seam coal gave efficiencies of 62 and 65 percent, while the test of Illinois No. 6 gave 72 percent. The lower values are considered more representative of the small experimental producer because heat losses to the cooling water are greater than would be experienced with a large producer operated continuously

at steady conditions. Heat loss to the water-cooled stirrer is negligible
because its surface area is relatively small, but the 6 to 10 percent
loss to the water-cooled wall is 2 to 3 times greater than normal for
a large producer.

Steam: Coal Ratio

The addition of steam has two very important functions: To improve
the cold efficiency and to control clinkering of the ash. The decomposition
of steam by reaction with carbon is strongly endothermic; hence, some of
the sensible heat, which is lost when the gas is used cold, is converted
into potential heat in the gas in the form of hydrogen and carbon monoxide.
Enough steam must be added to prevent extensive clinker formation, which
clogs the grate and stops the fuel flow, but excess steam causes excessive
cooling and, passing through undecomposed, is wasted. Pittsburgh bed coal
has a strong tendency to form hard clinkers, and--by trial-- a steam-to-coal
weight ratio of 0.5 gave satisfactory results. Illinois No. 6 coal showed
no tendency to form clinkers with a steam-coal ratio of 0.4.

Gas Yields

Gas yields are dependent upon the air rate which limits the com-
bustion rate up to the point where fuel loss by entrainment in the gas
stream becomes excessive. The maximum air rate in these tests was the
full output of a compressor, 60,000 scfh. The gas yields at 80 psig
are 63-65 scf per lb of Pittsburgh seam coal and 55 for Illinois No. 6.
The smaller yield for the latter coal results in part from its lower
carbon content.

Some additional improvement in gas yield appears possible if air flow
is increased by adding more compressor capacity. Another approach would
be to add oxygen to the air blast. This method could be used with a
rotating grate discharging the ash as solids. The addition of oxygen
increases the temperature in the combustion zone, and excess steam must
be added to avoid clinker formation by ash fusion. When 100 percent oxygen
is used the preferred ash removal method is fluid slag. This would
require a hearth-type retort with oxygen admitted above the hearth through
tyeres. In either case, the higher gasification rate is not expected to
alter fuel bed characteristics, and stirring is expected to maintain
nonagglomerated bed conditions.

CONCLUSIONS

Short-time tests in gasifying strongly caking coal in a stirred-
bed indicate that continuous stirring of the fuel bed is necessary and
succeeds in maintaining the fuel flow. Nuclear density and level gages
supply the data needed to control operation of the producer and hold the
gas quality uniform. Further increase in gas production rate appears
possible by increasing the air blast rate. Some improvement in gas quality

may be obtained by closer control and optimization of operating conditions. Additional tests are needed to obtain data showing gas yields and quality at maximum coal feed rates and to determine operability using smaller sizes of coal.

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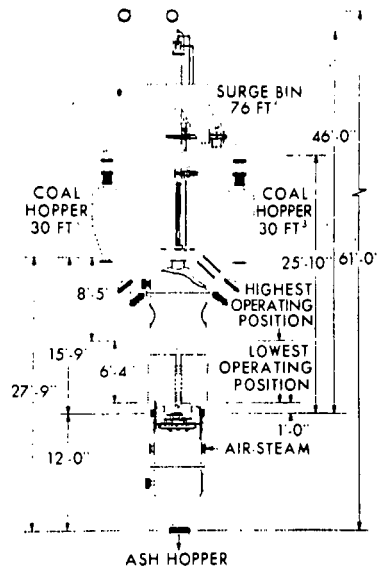
Figure 1. - Schematic drawing of gas producer

Figure 2. - View of gas producer
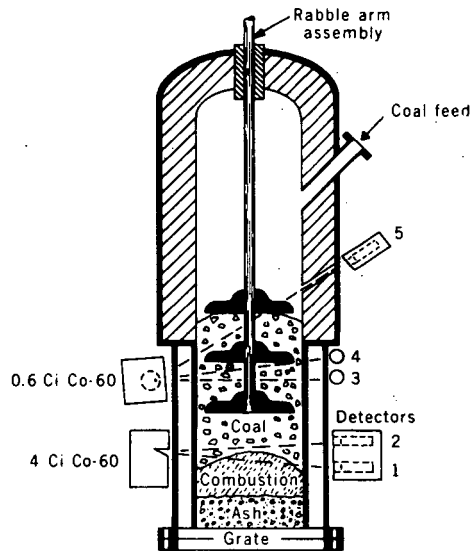
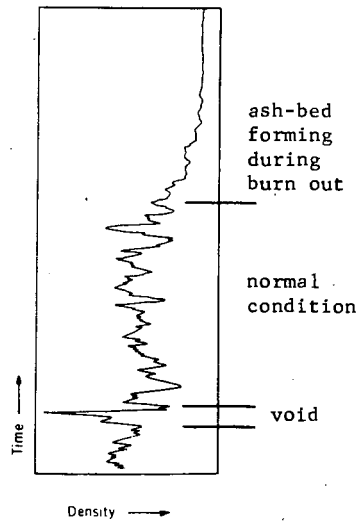Figure 3. - Nuclear density gages applied to gas producer

Figure 4. - Typical display and interpretation of density
measurements for the combustion zone

FLUIDIZATION PROPERTIES OF COAL CHAR

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INTRODUCTION

Coal reserves are vast and therefore will play an ever-increasing role in supplying our growing energy demands. However, because of predicted shortages of liquid and gaseous fuels and because of environmental considerations, future utilization will include the development of processes to desulfurize coal as well as to convert it to liquid and gaseous fuels that would also be desulfurized.

Thus, processes such as pyrolysis, desulfurization, gasification, hydrogasification, carbonization, as well as fluid-bed combustion, are being developed to allow coal to broaden its role in satisfying future energy and chemical demands. These processes have at least two features in common: First, they will likely utilize char, derived from coal, as a feed or a constituent in some reactor zone. Second, as a corollary of the particulate nature of char, the processes can use fluidized-bed reactors to take advantage of the improved heat and mass transfers, easier flow operations, good mixing, and ease of control. The design of reactor systems to carry out the above processes therefore requires knowledge of the fluidization properties of coal chars, as well as knowledge of reaction thermodynamics and kinetics.

This study was made to provide the necessary fluidization data for the design of fluid-bed hydrogasifiers of both laboratory and commercial scale. An excellent summarization of fluidization data for various materials appears in the literature.[1] Additional data on chars are also available.[2,3] However, the char we will be fluidizing is so different that use of the published data is precluded. The correlations presented here should allow extension of data to the design of any other fluid-bed reactor system using coal chars that have the same physical characteristics. Apparently, no underlined universal correlation has been developed to account perfectly for all the physical parameters involved in fluidization; thus, no single correlation now exists that suits all particulate materials regardless of constitution or especially particle shape. Separate correlations still appear to be needed for widely varied materials, as will be demonstrated here. The particular char used in these experiments was produced by rapid hydrogasification of raw high-volatile bituminous coal in a "free-fall" reactor. In our overall process concept to produce pipeline gas from coal, this lightly converted char (25 to 30 percent carbon conversion) would fall into a fluid-bed reactor for additional carbon conversion and for the removal of sufficient hydrogasification heat to maintain the reaction temperature at 1,600° to 1,700° F. The free-fall reactor system has been analyzed[4] and an integrated reactor system is now being designed to add a fluid-bed reactor below this dilute phase to allow both an experimental and an engineering evaluation of the overall process thus integrated.

EXPERIMENTAL

Material. The char used in this study was produced by concurrent injection of raw bituminous coal and hot hydrogen:methane (1:1) mixture into the top of a 5 ft X 3 in. id reactor at 1,000 psig and 1,650° F. Very rapid devolatilization, gasification of the volatiles, and partial hydrogasification of char all occur. The resultant char falls freely in the slowly down-flowing gas. Carbon conversion in the above reactor is from 25 to 30 percent.

Apparatus. The fluidization studies were conducted at atmospheric pressure with the majority of the tests in a plexiglass column of 3.69-inch id. The fluidizing gas was introduced at the bottom of the column through a porous sintered stainless steel distributor plate, 1/4 inch thick with an average pore size of 5 microns. Thinner, more porous distributors were avoided when found to induce more channeling in the fluid bed, especially at the reactor bottom. The bed depth was varied from almost 3 feet to a few inches with no appreciable change in fluidization properties. Two experiments were made in a two-inch id tube with static heights of approximately five feet to observe the fluidization behavior of this char at conditions anticipated for a bench-scale fluidized-bed char hydrogasifier. Gas rates and pressure drops were measured with the usual rotameters and manometers.

<div align="center">RESULTS</div>

Char Properties. A char property important to fluidization is the effective particle density. Because of the sudden heating in the dilute-phase hydrogasifier as described above, this char has a very large fraction of internal void space so that the density is much less than that of the feed coal or chars from slow carbonization. A photomicrograph of a typical char particle is shown in figure 1 to illustrate the internal structure of this char.

Because of the vesicularity of this char, the effective density of the char particles is not known directly, but can be calculated from bulk density and gas-flow pressure drop measurements made on packed beds according to the procedure recommended by Leva[5] using Ergun's correlations.[6]

As shown in figure 2, the particle densities measured in the above manner fall into two categories, depending on the particle diameter. For fine particles where $\bar{d}_p$ <.003 in., the effective particle density is about 26 lb/cu ft while for the coarser particles where $\bar{d}_p$ >.01 in., the particle density is about 16 lb/cu ft. For $.003 \leqslant d_p$ <.01 there seems to be a transitional region, although only one date point is included in this particle size range. The most likely explanation of this behavior is that below a certain particle diameter, the particles cannot be vesicular and therefore must have a higher effective solid density than larger particles which can contain many vesicules. This explanation is consistent with the microphotograph shown in figure 1 because the approximate size range of the vesicules is about equal to the particle diameter at which the density transition occurs. Therefore, because the vesicules seem to have a limiting minimum size, particles smaller than this minimum size are nonvesicular. Physical interpretation of this effect is possible. Volatile matter evolved inside small particles of rapidly heated coal can escape without forming a vesicule because of the shorter diffusional path, or because any "skin" of coal in its plastic phase is too thin and bursts before a sizable vesicule can form. The larger particles can have thicker, stronger plastic layers to contain the evolved volatile matter long enough to make vesicules, and the diffusional escape of gas is, of course, more restrained.

The bulk density dependence on mean particle size is shown in figure 3. The mean particle diameter is here defined, as recommended by Leva,[4] by $\frac{1}{\bar{d}_p} = \Sigma \frac{Xi}{d_{pi}}$. The

discontinuity of bulk density versus size occurs at the same particle diameter as was observed for the discontinuity of particle density versus size. However, the bulk density curve for the vesicular particles is slightly above the curve for the nonvesicular particles in spite of the higher solid density of the nonvesicular particles. The most likely explanation for this experimental observation is that the wider variety of particle shapes one finds in the larger particle size mixtures permits closer packing which more than compensates for the decreased solid density.

The smaller sized particles, being more spherical, apparently pack with a higher void space, causing a decrease in the bulk density.

Minimum Fluidization Velocity. The minimum fluidization velocity was found to depend on the properties of the solid and fluid in a way that permits use of the dimensionless Galileo number, N_{Ga} for correlation. A correlation that empirically fits the data was obtained by plotting log (N_{Re}) against log (N_{Ga}), as suggested by Wen and Yu.[1] The resulting correlation is shown in figure 4. An excellent straight line results for both narrow and wide particle size distributions when the mean particle diameter as defined above is used.

Some other salient features of the correlation presented in figure 4 are:
(1) Though self-consistent, our correlation using mean particle sizes does not agree with that of Wen and Yu.[1] (2) For narrow size distributions, as expected, it does not make much difference whether mean or maximum particle size is used; a self-consistent correlation occurs either way. (3) For the few points available for broad size distributions, attempted correlation is poor when maximum particle size, as recommended by Wen and Yu, is used instead of mean particle size.

In view of the above three features, it is recommended that average particle diameters be used with our correlation for predictive purposes with similar chars.

In general, the chars used in these studies require much larger velocities to fluidize than predicted by reference 1. The difference between the correlations is too great to attribute to inaccuracies in measuring ρ_s which is the least accurate term in the Galileo number. Examination of the char particles reveals the most likely reason for the deviation between correlations is the configuration or shape of the respective particles. A picture of some typical char particles in figure 5 shows that the "particles" consist of a sizable fraction of agglomerates of small particles and that these agglomerates can readily interlock. Thus, collisions between particles in the fluid bed will frequently result in the particles catching together which results, momentarily, in an increase in the effective particle diameter in the bed and the requirement of additional energy to move the combination or to unhook the particles. Because this additional energy can only be supplied through increased gas velocity, these particles fluidize at higher velocities than particles having identical properties except for less ability to interlock upon collision. In deciding whether to use the correlation based on these results or that based on more "regular" particles, it would probably be sufficient to examine the particles to see whether they contain shapes that can cause them to hook together and thereby require the use of higher fluidization velocities.

A summary of the fluidization data in given in table 1 to illustrate the range of particle sizes and bed heights used to derive the correlation in figure 4. The minimum fluidization velocity was obtained in the classical manner by plotting the experimentally measured bed pressure drop against the gas velocity, and defining the velocity at which ΔP just reaches $\Delta P = \dfrac{W_s}{A_T}$ as the minimum fluidization velocity.

A typical ΔP vs u curve is shown in figure 6a for a relatively narrow particle size range.

In general, fluidization was visibly smoother for narrow size fractions of particles. However, wide fractions of only coarser particles also behaved well. The most irregular fluidization was experienced with wide size fractions having appreciable concentrations of fine particles, leading to behavior illustrated in figure 6b,

TABLE 1. - Summary of fluidization data for coal char fluidized with CO_2; tube id = 3.69 (with exceptions noted below)

Particle size range, U. S. Sieve Series	Mean particle diameter, in.	Minimum fluidization velocity, ft/sec	Reynold's No. at minimum fluidization	Galileo number	Bed height at minimum fluidization, in.	Wt of char in bed, lb
6 x 8	0.113	1.42	151	3.6×10^5	9.0	0.248
8 x 12	0.08	1.06	80	1.2×10^5	11.0	0.339
12 x 14	0.061	0.74	43	6.24×10^4	8.2	0.280
14 x 20	0.044	0.57	23.8	2.5×10^4	10.8	0.410
20 x 40	0.025	0.37	8.8	5.15×10^3	10.6	0.480
40 x 60	0.013	0.14	1.8	810	3.7	0.209
60 x 80	0.0084	0.11	0.87	254	2.6	0.183
80 x 120	0.0059	0.085	0.48	101	2.0	0.163
120 x 200	0.0039	0.059	0.21	28.6	11.6	0.789
200 x 325	0.0022	0.040	0.080	6.02	13.0	1.51
1/6 x 40	0.065	1.23	75	1.34×10^5	24.0	0.546
1/6 x 40	0.071	1.27	85	1.57×10^5	30.0	0.825
[1,2]/100% -14, 34% -325	0.0052	0.05	0.25	61	47.0	1.0
[1,2,3]/100% -14, 34% -325	0.0052	0.03	0.076	26	47.0	1.0

[1]/ Data given by solid points on figure 4.
[2]/ Tube id = 2 inches.
[3]/ Fluidized with N_2.

where precise definition of U_{mf} was difficult. Appreciable channelling and segregation was noted with the particle size range illustrated in figure 6b, but this could be overcome by going to velocities considerably higher than U_{mf} where performance stabilized even with the relatively high L/D ratio used in this test.

Design Considerations. To design a fluid-bed chemical reactor, it is generally not sufficient to know only U_{mf} because one must operate at a velocity sufficient to give vigorous mixing and prevent the segregation of particles. With the narrow particle size ranges, it was found that a gas velocity twice that of U_{mf} was sufficient to provide good mixing and to prevent segregation. However, as mentioned above, for the wide particle size distribution (100 percent -14 to 34 percent -325) in table 1, it was necessary to operate at $U/U_{mf} > 7$ before complete mixing was achieved.

To calculate the mean solids residence time in a fluid-bed reactor requires knowing the amount of bed expansion as a function of operating conditions. The data below indicate that the expansion ratio depends on the mean particle size as well as U/U_{mf}. A summary of experimental results is shown in figure 7a and 7b where L/L_{mf} is plotted against U/U_{mf} for various particle sizes. As the mean particle size decreases, so does the bed expansion ratio for a fixed U/U_{mf} until d_p reaches about 0.025 in., below which further reductions in particle size do not change the expansion behavior. For wide particle size ranges, the previously defined mean diameter determines the bed expansion as is indicated in figure 7b where expansion ratios for fluid beds having char particles from 14 to 325 mesh are plotted. Also in figure 7b, data are shown for narrower particle size distribution tests made with particle diameters below .025 inch.

CONCLUSIONS

The minimum fluidization velocity of coal chars, physically similar to those studied here, can be predicted for various particle sizes and gas conditions by computing the Galileo number and fitting it to the correlation developed here. To get the Galileo number requires knowledge of mean particle size, particle density, and gas density and viscosity at conditions for the desired system. The correlation applies to both narrow and wide particle size distributions and over a range of "Reynolds numbers at minimum fluidization" from less than 0.1 to over 100. The minimum fluidization velocity predicted by our experimental correlation is considerably higher than one would calculate from existing theory and from other correlations because of small agglomerates in the char which cause entanglement and thus require an additional energy input to either separate them or fluidize the more massive combination. Correlations are also presented for fluid-bed expansion that indicate that the mean particle size is the main factor in determining how bed expansion will vary with gas velocity.

NOTATION

U = superficial gas velocity, ft/sec

U_{mf} = superficial gas velocity, at minimum fluidization, ft/sec

ρ_F = density of fluid phase, lb/cu ft

ρ_s = effective density of solid phase, lb/cu ft

ρ_b = bulk density of bed, lb/cu ft

d_{pi} = particle diameter, ft

Xi = weight fraction of particles having diameter d_{pi}

d_p = mean particle diameter, ft

μ = gas viscosity, lb/ft-sec

W_s = weight of solid in bed, lb

A_T = cross sectional area of tube, sq ft

ΔP = pressure drop across bed, lb/sq ft

L = bed height, ft

Lmf = bed height at minimum fluidization, ft

D_T = tube diameter, ft

g = acceleration of gravity, ft/sec^2

N_{Ga} = Galileo number = $d_p \rho_f (\rho_s - \rho_f) g \mu^{-2}$

(N_{Re}) = Reynold's number at minimum fluidization

$\qquad$ = $d_p U_{mf} \; \rho_f \mu^{-1}$

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ACKNOWLEDGEMENTS

FIGURE 1.- Cross Section of Typical Char Particle
at 220 Magnification.
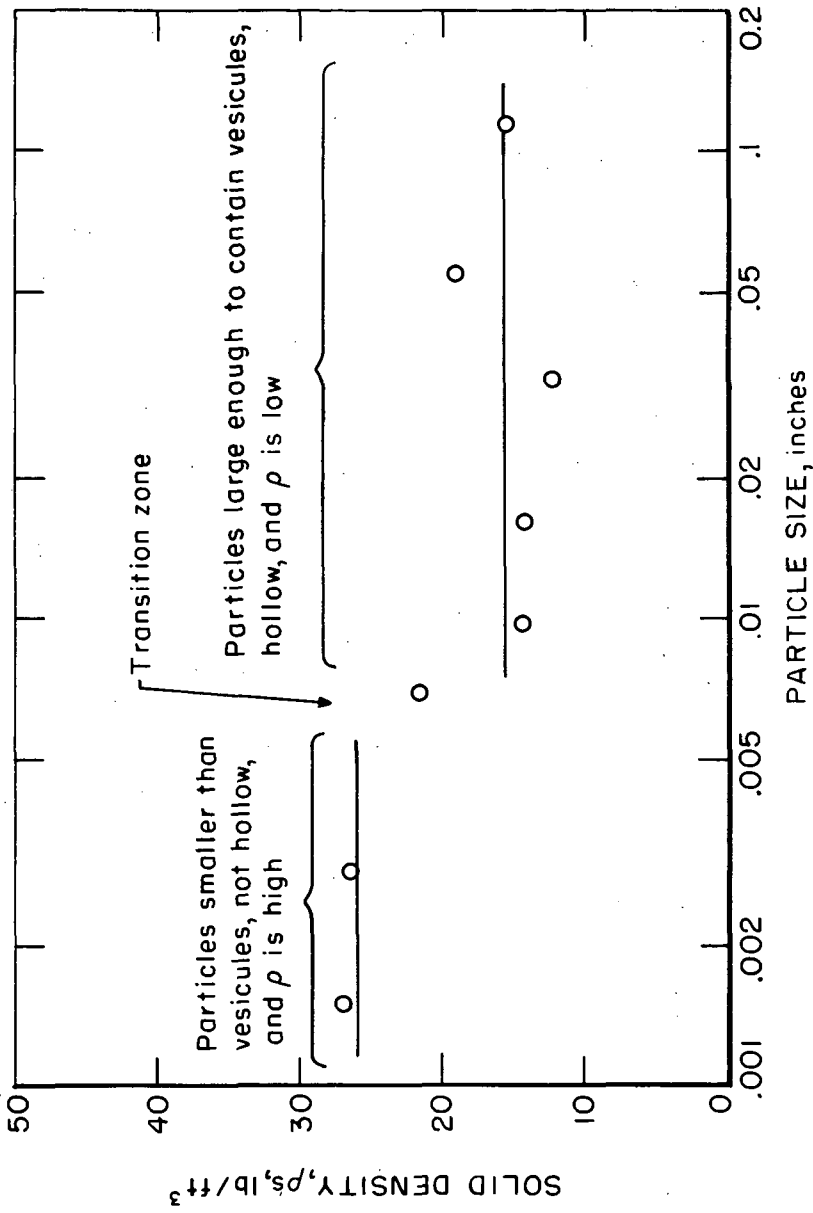
FIGURE 2.- Particle Density vs Particle Size of Char.

L-12230
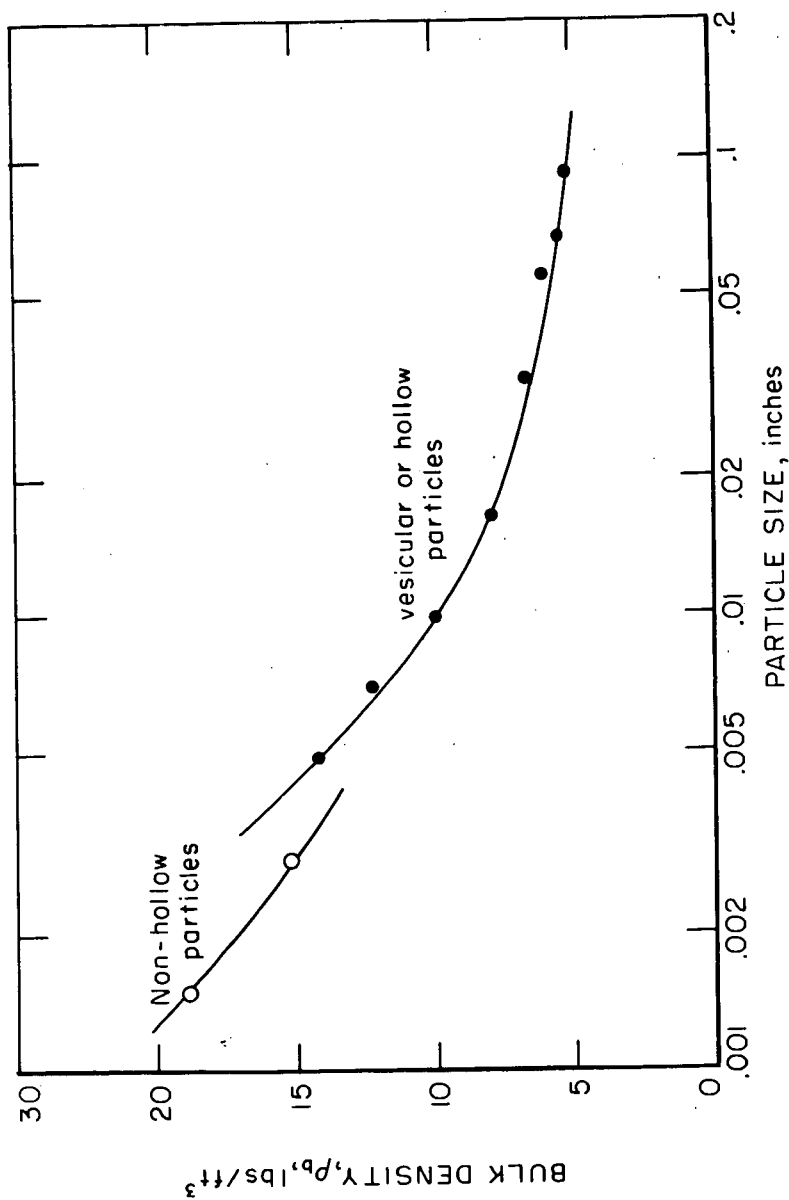
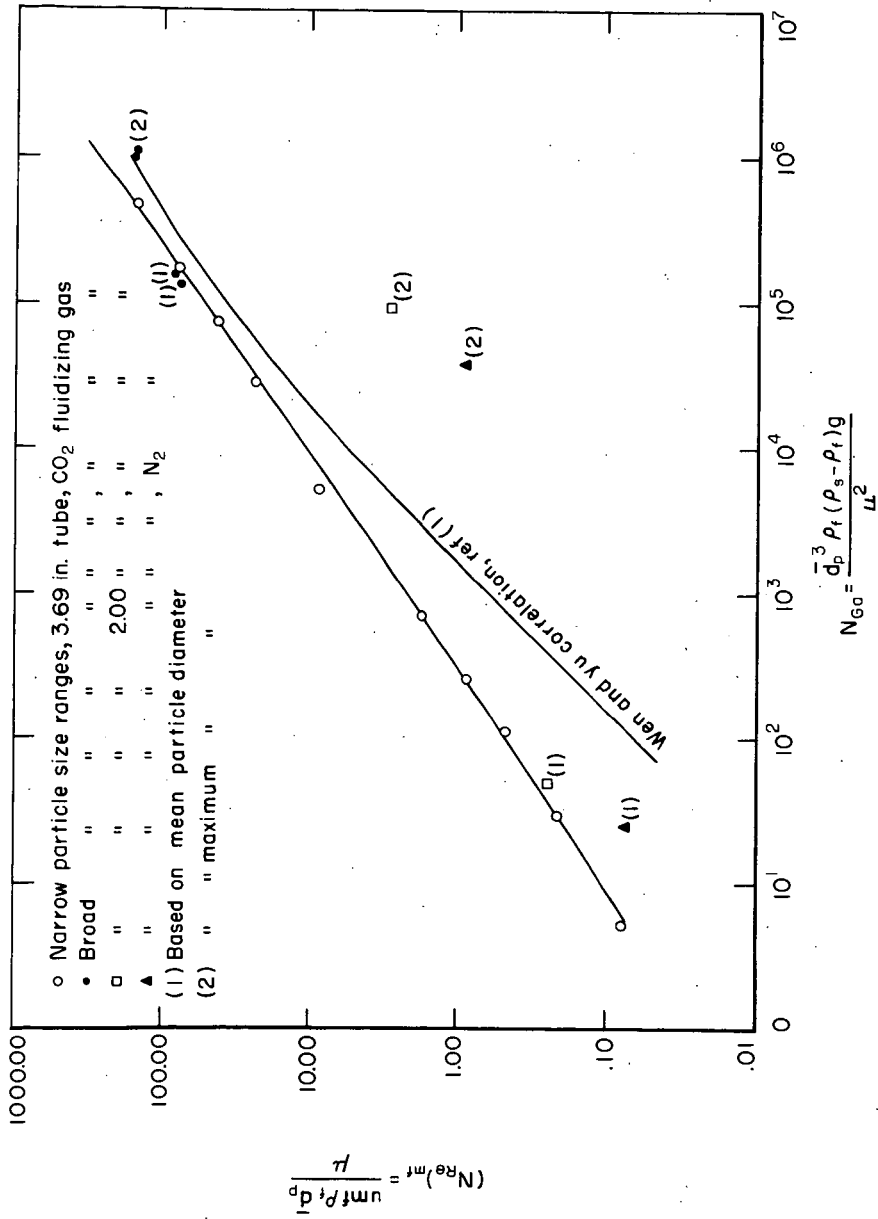FIGURE 3.- Bulk Density vs Particle Size of Char.
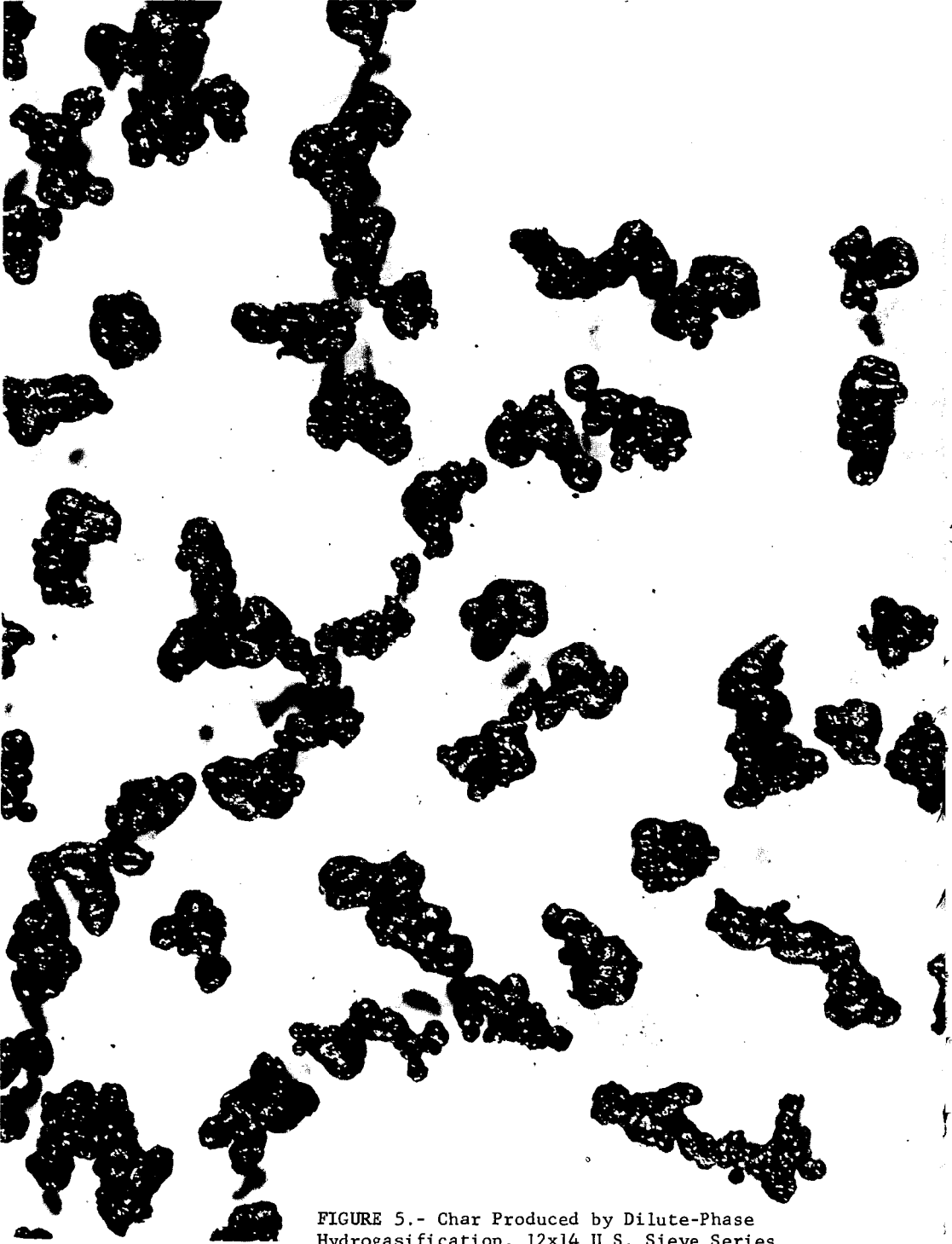
L-12231

FIGURE 4.- Fluidization Correlation.

L-12232

FIGURE 5.- Char Produced by Dilute-Phase
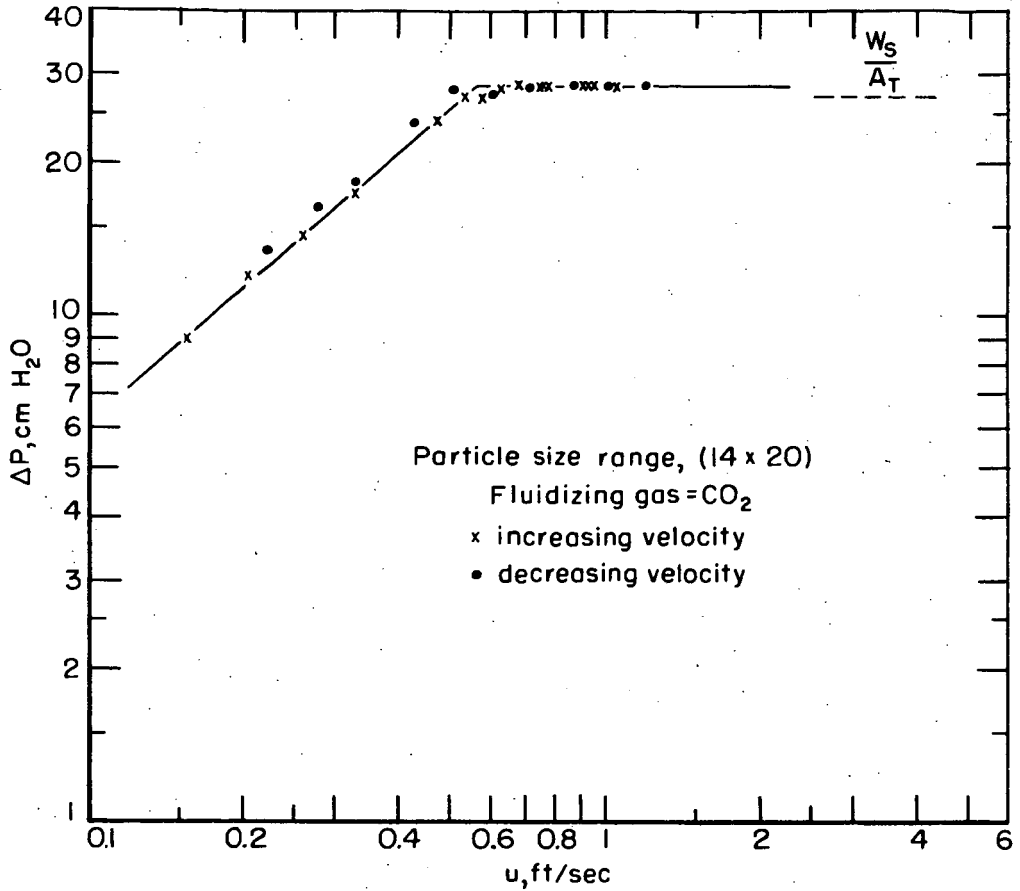Hydrogasification, 12x14 U.S. Sieve Series

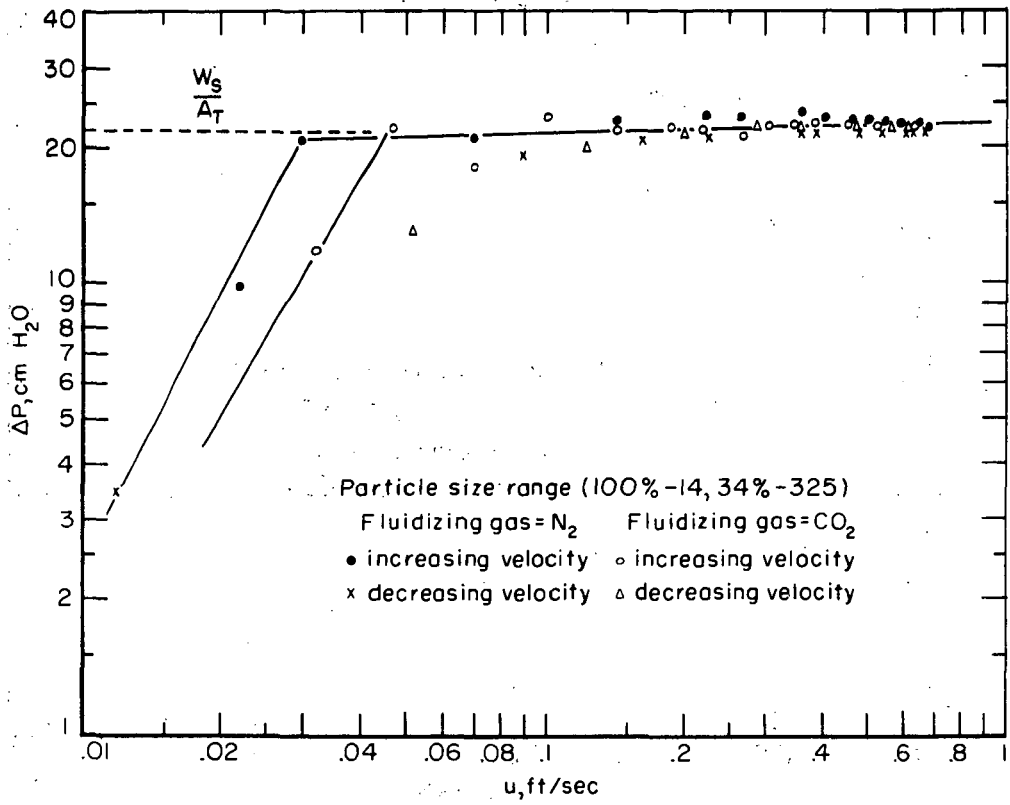FIGURE 6a.- Typical Pressure Drop-Velocity Curve, Narrow Particle Size Range.

L-12233

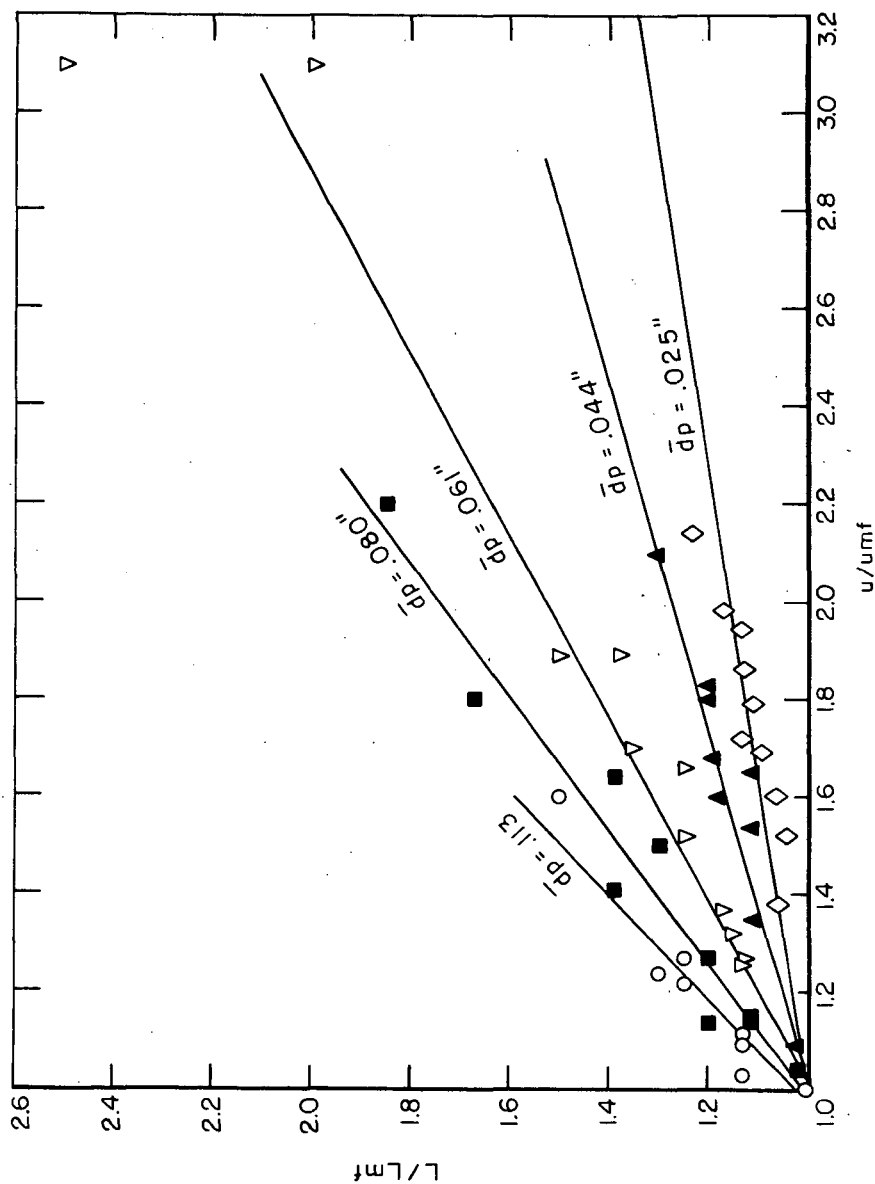FIGURE 6b.- Typical Pressure Drop-Velocity Curve,
            Broad Particle Size Range.

L-12234

FIGURE 7a.- Effect of d̄p and u/Umf on Bed Expansion.

L-12235
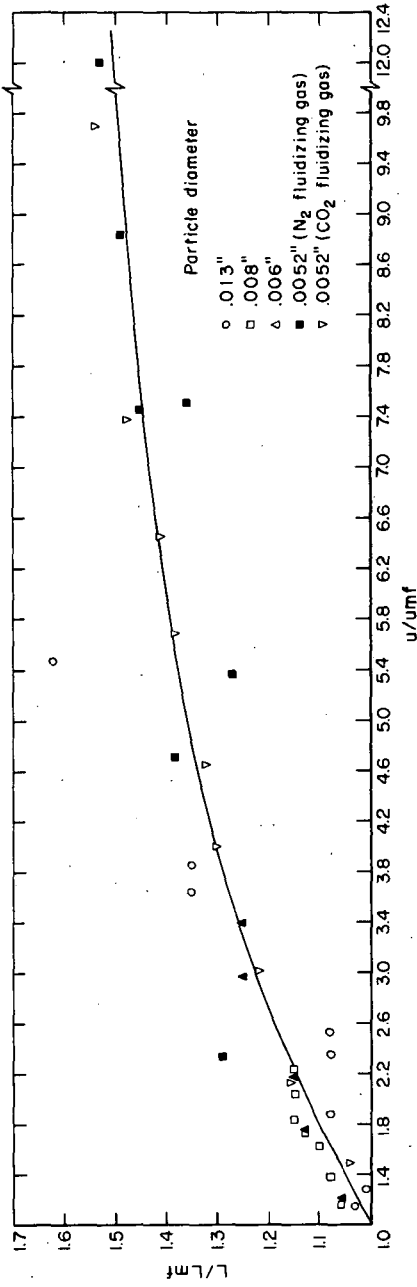
FIGURE 7b. - Effect of u/U_{mf} on Bed Expansion for $\bar{r}_p < .025$.

L-12236

STUDIES ON THE PRODUCTION OF CARBON BLACK FROM COAL

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ABSTRACT

Earlier work revealed that high volatile, vitrain rich and low ash coals could be converted to carbon black equivalent to thermal grade carbon black of commerce. In the present paper which is an extension of the earlier work some other coals have been used. It has been shown that by changing the reactor design and reaction variables the carbon black obtained is found to have properties of rubber and tyre grade products. Properties and compounding tests of the carbon black in rubber mix have been carried out and compared with those of standard products. A cost economics of the carbon black obtained from coal has also been discussed.

Coal has traditionally been used as fuels and there are very few studies describing the nonfuel use of coals. Johnson and co-workers (1,2) were the first to report on the conversion of high volatile coals into thermal black. They obtained the black as a byproduct in the production of hydrocyanic acid from coal. In an earlier paper (3) the present authors have described the production of thermal black from Assam coal. They adopted a transport reactor very much different to that used by the earlier workers and their main product was carbon black.

But thermal blacks find very limited use. The major demand is for rubber grade black for use in the tyre industry. The present paper describes studies on obtaining improved grades of carbon black from three bituminous coals of India. The paper also describes the techno-economic aspects of producing carbon black from coals.

Equipment and the process:

While the apparatus in the present studies are similar to that described earlier (3), the reactor, however, was designed to handle 70 lb./hour of coal. The inner wall of the reactor was made of 9 in. thick high alumina bricks followed by a 6 in. layer of insulation brick followed by a M.S. sheet. The reactor was 12 ft. high and 1 ft. in diameter.

A typical run was carried out as follows:-

Crushed coal (40 per cent through 200 BS) fed to the reactor through a rotary feeder is conveyed through the transport reactor by the carrier gas air. The reactor is initially heated to 500 - 600°C by burning low ash firewood. When the temperature becomes steady at the above temperature, the charging of coal is started. Partial combustion of the coal raises the temperature to 1300°C maximum. The air to coal ratio of the charges is kept constant at 65 cu.ft./lb. and the time of residence of the coal particles in the reactor is about 1.3 seconds. The carbon black is collected in the temperature range of 1200 - 1300°C. The char is separated from carbon black in a stainless steel cyclone separator. The carbon is recovered by first scrubbing it with water in the scrubber and then by electrostatic precipitation.

The product is then pelletised and dried at 400 - 450°C. Strict control of air/coal ratio and feed rate are important as changes may lead to existence of uncracked tar in the carbon black and deteriorate its property or lead to excessive combustion and give lower yield of the product. Figure 1 shows the unit for producing carbon black from coal.

Experimental results and discussion -

The work was carried out with three different coals of India formed under widely varying geological conditions. Table I shows the analysis of the coals used in the study.

Though they are all high volatile low rank coals, they differ in their caking characteristics and ash content. Sulphur content of the third coal is much lower than the two other coals. Reaction conditions were maintained identical and average test result of two or three runs has been shown. Table II shows the analysis of carbon blacks.

Figure 2 shows that the yield of carbon black bears a direct relationship with the hydrogen content of the coal. The Russians also obtained such relations (4), but details of their process are not available. The formation of carbon black from coal initially involves decomposition of coal to tar and gaseous hydrocarbons which ultimately undergoes dehydrogenation and aggregation to form carbon black. Theories on the carbon black formation differ mainly in the route and order in which these two essential changes occur (5).

Coals containing high mineral matter give a higher ash in the product. Consequently compounding tests have been carried out with only low ash carbon black produced from Baragolai colliery coal, Upper Assam. Figure 3 shows the electron micrographs of carbon black produced from Baragolai colliery coal (a) and HAF black(b). The study was carried out using a magnification of 52700 to determine the average particle size and the structure of the black. It is observed that carbon black obtained from coal is of high structure and is different from thermal black, which is also supported by its compounding test. The size of these particles varies from 200 - 800° A comparable with HAF blacks having particle size varying between 200 - 800°A. However, the frequency of distribution of bigger particles in the HAF black is much less than those in carbon black prepared from coal.

Figure 4 shows the x-ray pattern of carbon blacks obtained from (1) Baragolai colliery coal, (2) Pench Valley coal, and (3) Standard Sample HAF. It may be observed that the carbon blacks obtained from coals show bands of mineral matters, especially quartz, which is more prominent in the case of sample 2 obtained from Pench Valley coal containing higher percentage of mineral matter. The carbon band of samples 1 and 2 differ in intensity from that of number 3 as the formation of the latter takes place at temperature higher than those of 1 and 2. This may be attributed to the randomness in the degree of orientation of carbon particles in samples 1 and 2 whose formation takes place at lower temperature than that of sample 3.

Properties of the carbon black obtained from Baragolai colliery coal, Assam have been compared with two commercial carbon blacks in Table III.

Acetone extract which indicates the presence of uncracked tar in the product is slightly higher than those of the standard blacks. Iodine absorption values are greater than even HAF blacks. Surface area has been calculated from iodine absorption values (6), using the relationship $SDd = 60,000$, where S = surface area, D = density, d = particle size. D.B.P. absorption is very nearly equal to the HAF blacks but greater than the SRF blacks. pH of the product is in the mild acidic range which may be attributed to the nature of starting material.

Table IV shows the physical properties of rubber compounded with carbon black obtained from Baragolai colliery coal and compared with those compounded with a standard SRF black. Mooney viscosity which is an important property of the polymer mix is dependant mainly on the carbon black structure and loading in most polymers. In a 50 phr sample Mooney viscosity @ 100°C is found to be higher than the SRF blacks and Mooney scorch time about 0.64 times that of SRF blacks.

Rate of cure of the compound loaded with carbon black derived from coal is faster than compounds loaded with SRF blacks.

Physical properties like tensile strength, elongation at break and 300% modulus of the compound loaded with carbon black from coal are in line with SRF blacks, which shows it can substitute SRF blacks.

Cost economics:

A cost study of carbon black produced from coal revealed that the cost of production is $ 70 per ton on the basis of 20 per cent conversion of coal to carbon black and price of coal being taken $ 7.5 per ton. The price of char produced in the process (about 30 per cent) is taken as $ 10.00 per ton. This is quite promising as the market price of carbon black in India varies from $ 225 - 325 per ton.

Conclusion -

By changing the design and material of construction and altering the reaction variables the quality of the carbon black obtained from coal can be considerably improved to approach those of the rubber grade product. Some of the properties are similar to those of HAF. Though the physical properties like tensile strength, elongation at break and 300 per cent modulus of the compound loaded with carbon black from coal are lower than the compounds loaded with HAF blacks, they are in line with the SRF blacks. Experiments are still underway to produce HAF black.

Acknowledgement:

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Table I - Analysis of the coals used for production of carbon black.

Details of the coals	Ash %	Mois-ture %	V.M. % DAF	Fixed Carbon %	C % DAF	H % DAF	S % DAF	N+O by diff. % DAF	Caking proper-ties
1. Baragolai colliery, Upper Assam	2.7	1.8	42.3	57.7	81.4	5.6	2.5	10.5	Highly caking.
2. Garo Hill, Lower Assam	6.5	1.4	49.5	50.5	80.3	5.8	2.5	11.4	Weakly caking.
3. Pench valley, M.P.	23.4	4.5	35.0	65.0	82.5	5.2	0.7	11.6	Non-caking.

Table II - Analysis of carbon blacks produced from different coals under reaction conditions.

Coal feed rate - 70 lb./hr.

Air/coal ratio - 65 cu. ft./lb.

Temperature range of collection of carbon black - 1200 - 1300°C.

Carbon black obtained from	Yield %	Ash %	Moisture %	V.M. %	F.C. %	C %	H %	S %	N+O (by diff.)
1. Baragolai colliery, Upper Assam.	20	1.3	2.0	2.2	94.5	95.8	0.7	1.6	1.9
2. Garo Hill, Lower Assam.	22	4.2	2.5	2.6	90.7	95.4	0.8	1.7	2.1
3. Pench Valley, M.P.	14.4	18.3	1.4	3.8	76.5	94.7	0.7	0.4	4.1

Table III - Properties of carbon black obtained from Assam coal compared with those of commercial carbon black.

Carbon black obtained from	Acetone extract %	Iodine absorption values mg./g.	D.B.P. absorption values ml/100g.	Particle size, °A	Surface area M²/g.	pH
1. Baragolai colliery coal, Upper Assam.	1.9	89	106	200 - 800	68	6.5 to 7.0
2. SRF black	0.4	30	70 - 80	600 - 800	25 - 30	6.0 - 6.5
3. HAF black	0.5	82	103±6	200 - 800	74 -100	8.0 - 9.0

Table IV - Physical properties of rubber compounded with *ACB and SRF.

Compound No.	50 phr SRF	50 phr ACB sample	60 phr ACB sample	70 phr ACB sample
Mooney viscosity (ML4) @ 100°C	63.5	74.5	82.0	95.5
Mooney scorch (MS) Time @ 127°C.	32'-43"	21'-30"	20'-25"	18'-30"
Cure Index (RC) @ 127°C	6'-0"	5'-0"	4'-40"	4'-30"
Time of cure @ 145°C	15' 25' 35'	15' 25' 35'	15' 25' 35'	15' 25' 35'
Tensile strength lb./sq. ft.	2946 2974 3060	2803 2817 2960	2789 2960 2760	2660 2617 2560
Elongation at break %	670 396 385	530 400 400	500 385 350	400 300 290
300% Modulus lb./cu.ft.	1217 2174 2230	1459 1945 1973	1730 2160 2274	2074 2560 -
Shore hardness A$_2$	61 65 65	68 70 70	70 71 71	77 79 79

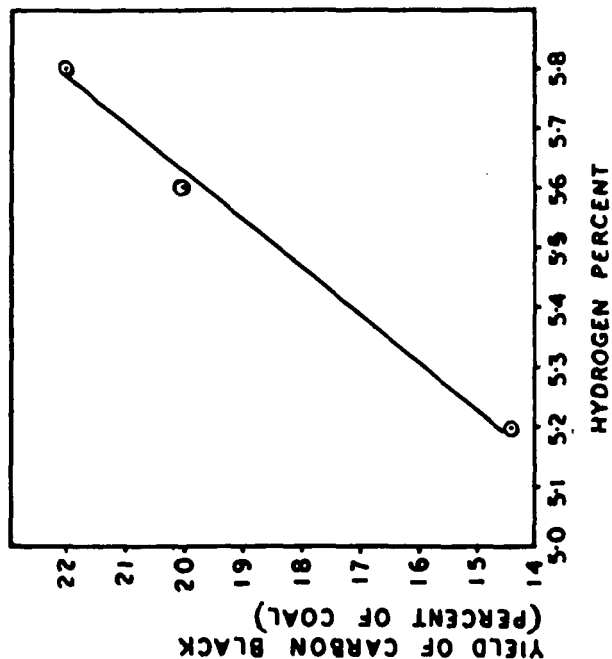*ACB indicates carbon black obtained from Baragolai colliery coal, Upper Assam.

FIGURE-2. RELATIONSHIP BETWEEN
HYDROGEN CONTENT OF COAL
AND YIELD OF CARBON BLACK
FROM IT.

Figure 2. Relationship between hydrogen
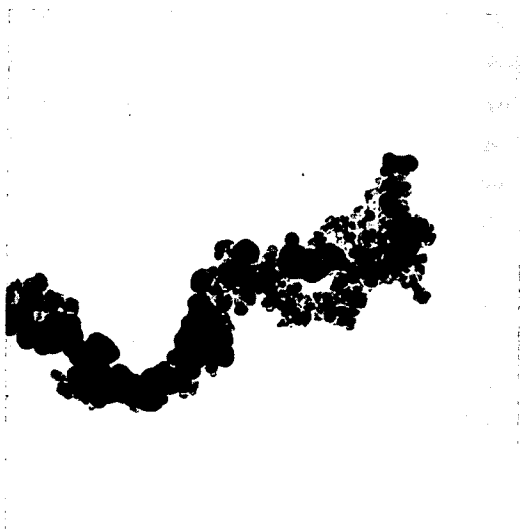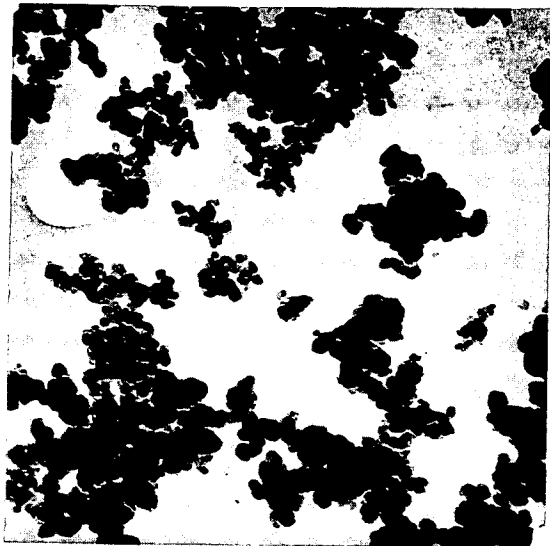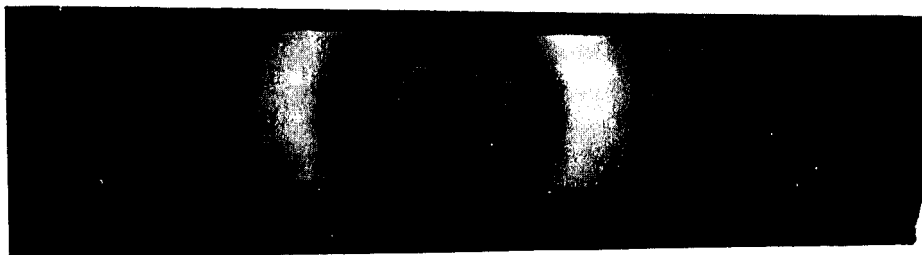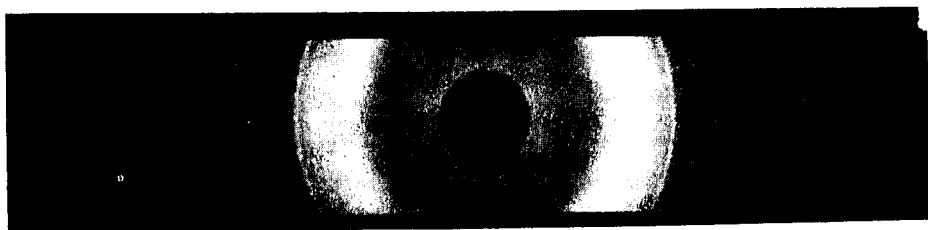content of coal and yield of carbon
black from it.
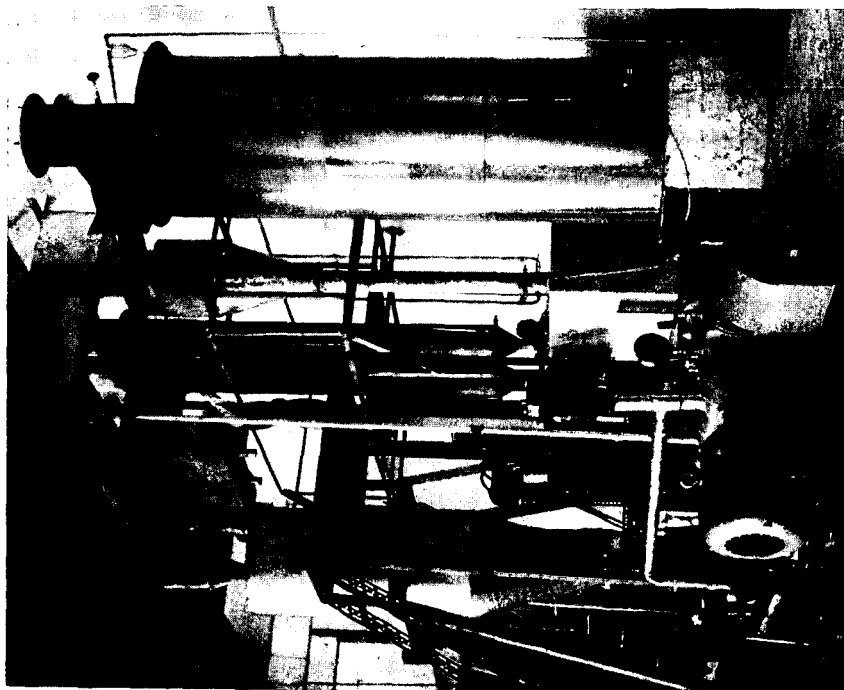
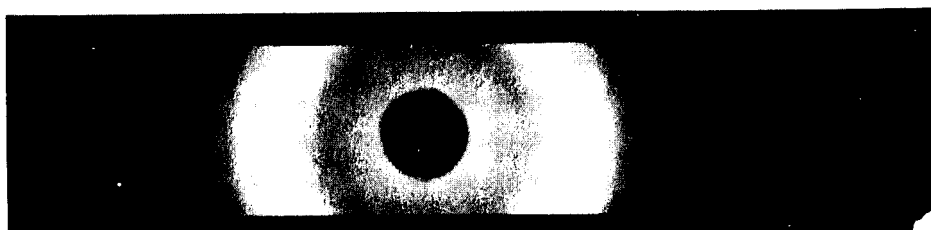Figure 1. The unit for the production of
carbon black from coal.

Figure 3. Electron micrograph of carbon blacks produced from (a) Baragolai colliery coal, Upper Assam, (b) HAF black (magnification 52700 times).

(1)

(2)

(3)

Figure 4. X-ray diffraction patterns of the carbon
blacks obtained from (1) Baragolai colliery
coal, Upper Assam (2) Pench Valley coal,
M.P., (3) HAF black.